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NATIONAL STATUS AND TRENDS, MUSSEL WATCH PROGRAM

An Assessment of Contaminants of Emerging Concern in the Gulf of Maine

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NOAA TECHNICAL MEMORANDUM NOS NCCOS 291

NOAA NCCOS Monitoring and Assessment Branch

Front Page Images (Clockwise): Boston Harbor. Credit: NOAA; Mytilus species. Credit: NOAA; Gulf of Maine. Credit: NOAA; Mytilus species. Credit: NOAA

Back Page Image: Gulf of Maine. Credit: NOAA

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Contents

Abbreviations

AFR	Alternative flame retardant
AP	Alkylphenol compound
BFR	Polybrominated flame retardant
CSO	Combined sewer outfall
CUP	Current-use pesticide
EPA	Environmental Protection Agency
g	gram
Gw	Gulfwatch Program
HUC	Hydrological Unit Code
MA	Massachusetts
MDL	Method detection limit
ME	Maine
MRLC	Multi-Resolution Land Characteristics
MWP	Mussel Watch Program
NCCOS	National Centers for Coastal Ocean Science
ng	nanogram
NH	New Hampshire
NS	Nova Scotia
NS&T	National Status & Trends
NLCD	National Land Cover Database
PBB	Polybrominated biphenyl
PBDE	Polybrominated diphenyl ether
PFAS	Per-and polyfluoroalkyl substances
PPCP	Pharmaceutical and personal care product
USGS	United States Geological Survey
ww	wet weight
WWTP	Wastewater Treatment Plant

Figures

Figures (cont.)

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Tables (cont.)

Executive Summary

In collaboration with the Gulf of Maine Council on the Marine Environment (GOMC) Gulfwatch Program, the National Oceanic and Atmospheric Administration (NOAA) Mussel Watch Program (MWP) conducted an assessment of the presence, distribution, and concentrations of contaminants of emerging concern (CECs) in bivalves in the Gulf of Maine's coastal waters. Like the national MWP, the Gulfwatch monitoring program utilizes a sentinel-based monitoring approach by collecting and analyzing bivalves as surrogates for coastal water pollution. A total of 52 composited blue mussel tissue samples were analyzed for this study, from a combined 41 monitoring sites. Following modified national MWP standard protocols (Apeti et al., 2012), 37 samples were collected in 2016 from selected MWP and Gulfwatch sites. The remainder were frozen Gulfwatch Program samples previously collected in 2015. Monitoring sites were located across four jurisdictions of the Gulf of Maine including, Maine, Massachusetts, New Hampshire, and Nova Scotia (Canada). The mussel samples were measured for a total of 249 individual CEC compounds, including 4 alkylphenol compounds (APs), 9 alternative flame retardants (AFRs), 33 current-use pesticides (CUPs), 12 per- and polyfluoroalkyl substances (PFASs), 121 pharmaceutical and personal care products (PPCPs), and 70 polybrominated flame retardants (BFRs) such as polybrominated biphenyls (PBBs) and polybrominated diphenyl ethers (PBDEs). Many of these compounds and their metabolites are shown to have estrogenic endocrine-disrupting effects, cause developmental problems in animals, or are suspected carcinogens.

The results indicated that many CECs are present in the Gulf of Maine's coastal waters and some of these chemical are found at various concentrations in coastal bivalves found in the region.

- Three of the four AP contaminants, 4-nonylphenol mono-ethoxylate (NP1E0), 4-nonylphenol di-ethoxylate (NP2E0), and 4-n-octylphenol (4-n-OP) were found above detection limits in mussels from different locations across the Gulf of Maine particularly in ME and NH. A maximum concentration of 16.5 ng/g wet weight (ww) was recorded at the South Mill Pond (NHSM) site in NH for 4-NP1EO.

- Two of the nine AFR contaminants, 2-ethylhexyl tetrabromobenzoate (TBB) and 2-ethylhexyl 3,4,5,6-tetrabromophthalate (TBPH), were found at monitoring sites in MA and ME, but more frequently in MA than ME. A maximum concentration of 3.27 ng/g ww for TBB was recorded in the mussel sample from the Deer Island (MADI) site in MA. TBPH was found at a concentration of 0.73 ng/g ww in mussels from Kennebec Perkins Island (MEKN) site in ME.

- Among the 33 CUPs tested, none were found above detection limits in the Gulf of Maine.

- Three of the 12 PFASs, perfluorooctanoate (PFOA), perfluorooctane sulfonate (PFOS), and perfluorooctane sulfonamide (PFOSA) were found across the Gulf of Maine. The most frequently detected of the PFASs, PFOSA, was detected at approximately 40% of the stations in the Gulf of Maine. PFOSA was found at 16 monitoring sites across MA, NH and ME and a maximum concentration of 5.46 ng/g ww was recorded for PFOSA at the Stroudwater-Force Portland Harbor (MEPH) site in Maine. PFOS and PFOA were respectively found at the Neponset River (MANR) site in MA with a concentration of 0.60 ng/g ww, and at the Boothbay Harbor (MEBB) site in ME with a concentration of 0.35 ng/g ww.

- Of the 121 PPCPs measured, 31 contaminants were detected across the study area. Within the Nova Scotia jurisdiction in Canada, only norgestrel (a birth control medication) was detected and

only at the Annapolis Basin Digby (NSDI) site. The most frequently detected PPCPs in the Gulf of Maine were, DEET, an insect repellant with a Gulf-wide detection frequency of 87.5% (of the sites), followed by sertraline, an antidepressant drug (42.5%), diphenhydramine, an antihistamine drug (30%), ranitidine, a gastroesophageal and heartburn drug (22.5%), and triamterene, a diuretic drug (12.5%). 17β-estradiol, a steroidal estrogen, miconazole, an antifungal medication, and propranolol, a hypertension and heart rhythm disorder drug, were also detected at two sites each Gulf-wide. Meprobamate, a sedative drug used for insomnia and psychiatric anxiety, and caffeine were infrequently detected, however they were found at concentrations of 59.4 and 57.7 ng/g ww respectively at the Boston Harbor Brewster Island (MABI) and Salem Harbor Folger Point (SHFP) sites in MA. Moreover, metprolol and propranolol, which are both used to treat angina and hypertension, were detected respectively at 46.7 and 42.6 ng/g ww in mussel tissues from the sites Royal River (MERY) in ME and Piscataqua River Dover Point (NHDP) in NH.

- Twelve of the 70 BFR contaminants were detected Gulf-wide. None of the PBBs were detected. The most frequently detected PBDEs were the congeners PBDE-47 found at 80.5% of the sites, followed by PBDE-99 (63.4%), PBDE-71/49 (58.5%), PBDE-119 (53.66%) and PBDE-77 (48.8%). The BFR contaminants were ubiquitous in the study area from Cape Cod to the Bay of Fundy in Nova Scotia, Canada, but the concentrations detected were relatively low, just above the MDL values. The maximum concentrations found across the study area were recorded for the congener PBDE-209 found at 1.04 ng/g ww and 0.96 ng/g ww at the Merrimack River (MAME) and Cohassett (MACO) sites respectively in MA. The congener PBDE-71/49 was measured at 0.76 ng/g ww at the Stroudwater-Fore Portland Harbor (MEPH) site in ME, and the congener PBDE-77 was detected at a concentration of 0.67 ng/g ww in mussel sample from the Hampton-Seabrook Estuary (NHHS) site in NH. BFR contaminants were detected more frequently at sites located at the mouth of rivers such as the Merrimack River (MAME) and the Neponset River (MANR) sites and estuaries such as Weir River Estuary (MAWR) and Hampton-Seabrook Estuary (NHHS).

The results indicated that CECs are present at various degrees in coastal waters of the Gulf of Maine and they are being observed at various concentrations in coastal bivalves. The detection of at least two CEC compounds at every monitoring location highlighted the presence of these contaminants in the coastal zone throughout the four Gulf of Maine jurisdictions studied in this report. However, the bioaccumulation of the CEC contaminants in organisms such as mussels is often compound and location dependent. That is, the presence and concentration of a specific contaminant are heavily influenced by its chemistry, sources, and fate and transport. Moreover, the distribution and magnitude of the CEC contaminants also depend on location and land-use types in watersheds adjacent to the monitoring location. Based on the land-use assessment in this study, CEC contaminants were located in coastal zones within all land-use categories, however, some contaminants were correlated with percent impervious surface or developed land-use. Wastewater treatment plants and outfalls may also be influencing the presence and concentration of some compounds.

Studies, such as this Gulf of Maine CEC assessment study, not only provide needed data and information for the national MWP, but also address crucial CEC monitoring data gaps for the Gulfwatch Program and support water quality data required by coastal resources managers as they develop effective long-term policies protecting ecosystem services provided by the coastal environment within this region.

A History and Description of the Mussel Watch Program

The national Mussel Watch Program (MWP), which began in 1986, was designed by the National Oceanic and Atmospheric Administration (NOAA) to monitor the nation's coastal waters for chemical contaminants and biological indicators of water quality. The MWP was established in response to a legislative mandate under Section 202 of Title II of the Marine Protection, Research and Sanctuaries Act (MPRSA) (33 USC 1442), which called on the Secretary of Commerce to, among other activities, initiate a continuous monitoring program. The MWP design is based on the periodic collection and analysis of bivalves (oysters and mussels) and sediment from a network of monitoring sites located throughout the nation's coastal zones. To date, NOAA's MWP is one of the longest running, continuous coastal monitoring programs.

The MWP monitoring sites are found along all of the US coastlines, including Alaska, the Great Lakes, Hawaii, and in territories such as Puerto Rico. Different target bivalve shellfish are used as sentinel species. Mussels and oysters are sessile organisms that filter and accumulate particles from water; thus, measuring contaminant levels in their tissue is a good indicator of local chemical contamination. The mussels (*Mytilus* species) are collected from the North Atlantic and Pacific coasts, oysters (*Crassostrea virginica*) from the mid-Atlantic (Delaware Bay) southward and along the Gulf Coast, and the invasive zebra and quagga mussels (*Dreissena* species) are collected from sites in the Great Lakes. Mangrove oysters (*Crassostrea rhizophorae*) are collected from Puerto Rico and Hawaiian oysters (*Dendostrea sandvicensis*) from Hawaii.

A fundamental challenge faced by any long-term environmental monitoring program is how (or whether) to evolve in response to changing conditions and drivers. In 2013, due to budgetary constraints, the National Centers for Coastal Ocean Science (NCCOS) undertook the task of re-designing the MWP, moving from a nationwide yearly monitoring approach to the rotating regional monitoring model that is currently employed. The regional approach allows the program to improve its presence in the coastal communities by increasing interaction with local stakeholders, integrating inputs from coastal resource managers, and providing specific data needs to help fill local data gaps. By making adaptive changes and leveraging regional partnerships, the program has increased its scientific relevance and reputation, and has evolved to include more than 300 monitoring sites (Figure 1) and nearly 600 chemical contaminants, which include metals, legacy organic compounds and chemicals of emerging concern (CECs).

The MWP provides unique data that is vital to evaluating the health of the nation's estuarine and coastal waters, particularly describing the levels of chemical contamination. The MWP dataset allows for temporal and spatial evaluation of regional and national changes in chemical distribution, including targeting CECs as their potential risks are identified. The program's long-term data supports the assessment of potential impacts of unforeseen events such as oil spills and hurricanes, as well as evaluating the effectiveness of regulations that ban toxic chemicals or support legislation such as the Clean Air and Clean Water Acts.

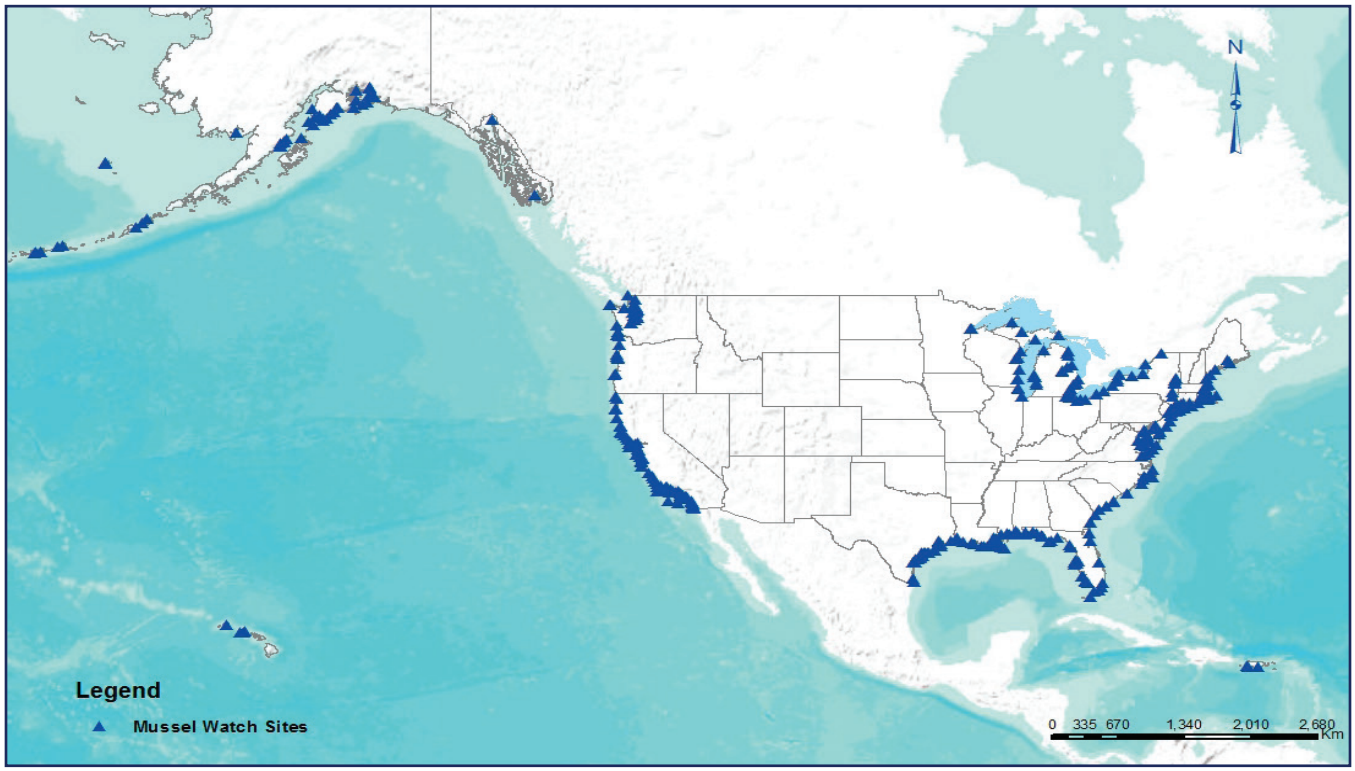


Figure 1. National Mussel Watch sites.

Mytilus species. Credit: NOAA

Dreissena species. Credit: NOAA

Crassostrea virginica bed in Charleston Harbor, SC. Credit: NOAA

A History and Description of the Gulfwatch Program

In order to protect water quality and sustain commercial uses in the Gulf of Maine, the *Agreement on the Conservation of the Marine Environment of the Gulf of Maine* was signed in December 1989 by the premiers of Nova Scotia and New Brunswick and the governors of Maine, New Hampshire and Massachusetts, thereby establishing the Gulf of Maine Council on the Marine Environment (GOMC). The overarching mission of the Council is to maintain and enhance the Gulf's marine ecosystem, its natural resources and environmental quality. The Monitoring Committee was a foundational part of the Council and developed a detailed monitoring plan for the Gulf of Maine (GOMC, 1991a), from which it eventually adopted the Gulfwatch Program as its main activity (GOMC, 1991b).

> *"It is the mission of the Gulf of Maine Environmental Quality Monitoring Program to provide environmental resource managers with information to support sustainable use of the Gulf and allow assessment and management risk to public and environmental health from current and potential threats."*
>
> Gulf of Maine Environmental Monitoring Plan (GOMC 1991a)

Since the inaugural year of the Gulfwatch Program in 1991 and throughout the history of the program, its mission, goals, hypotheses and objectives have changed with management needs, scientific findings and to reflect changes in resource availability. In support of the mission and as a first step towards meeting the desired goals and address a significant knowledge gap, the Gulfwatch Program was established to measure chemical contamination Gulf-wide (Figure 2). The GOMC Environmental Quality Monitoring Committee (EQMC) reviewed the results from the 2-year pilot project in early 1993 and decided to continue with the Gulfwatch Program with a modified, 9-year plan, (Sowles and Crawford, 1993) and the Council's Working Group concurred. One reason for modification of the existing program was the unexpected elevated contaminant concentrations at what were thought to be uncontaminated sites, especially in the southern Gulf of Maine, reflecting a lack of information on contamination levels and sources in many areas around the Gulf of Maine.

In the mid-2000s, the potential for expanding the program to include new yet related measurements, analyses and networking with other programs was being explored, yet the ongoing reality of limited available resources was finally acknowledged in a formal fashion with a new mission statement drafted by the committee in 2004 along with a new 12-Year Plan. In its most recent form, the Gulfwatch Program focus is:

> "*Using mussel tissue monitoring as a starting point, provide high quality and relevant data to allow for characterization of the condition of ecosystems in the Gulf of Maine for enhancing marine resource management and protecting public health.*"
> The new 12-Year Plan - 6/11/04

The program accomplishes this in part by conducting regional contaminant monitoring using the blue mussel, *Mytilus edulis*, as an indicator of habitat exposure to organic and inorganic contaminants and assessing the status and trends of chemical contaminants in coastal waters of the Gulf of Maine and Bay of Fundy. Rotational monitoring sites were to be sampled every six years and benchmark sites every two years. Some of the rotational sites were considered to be of heightened interest to managers and sampling increased to every three years for these targeted sites.

At a committee meeting prior to the 2007 season, significant new changes were adopted. Sampling would continue with replicate samples from each site, but replication was reduced from four to three, and contaminant analysis would only be done using a composite of the three replicate samples. The three replicate samples were to be archived. This freed up significant resources and allowed for much more frequent sampling. Thus, the frequency of sampling for the 12-Year Plan was doubled, so benchmark sites were sampled every year and rotational sites every three years. This design was used by the program through 2008.

In 2009, the number of sampling sites was further decreased and the timing of sampling was accelerated as part of a new 8-year plan for 2009-2016. Sampling supported by the GOMC ended in 2014, but samples were collected and stored in 2015, and in 2016 sampling in collaboration with NOAA Mussel Watch occurred and resulted in this report.

Please visit https://gulfofmaine.org/public/gulfwatch-contaminants-monitoring/ for more information.

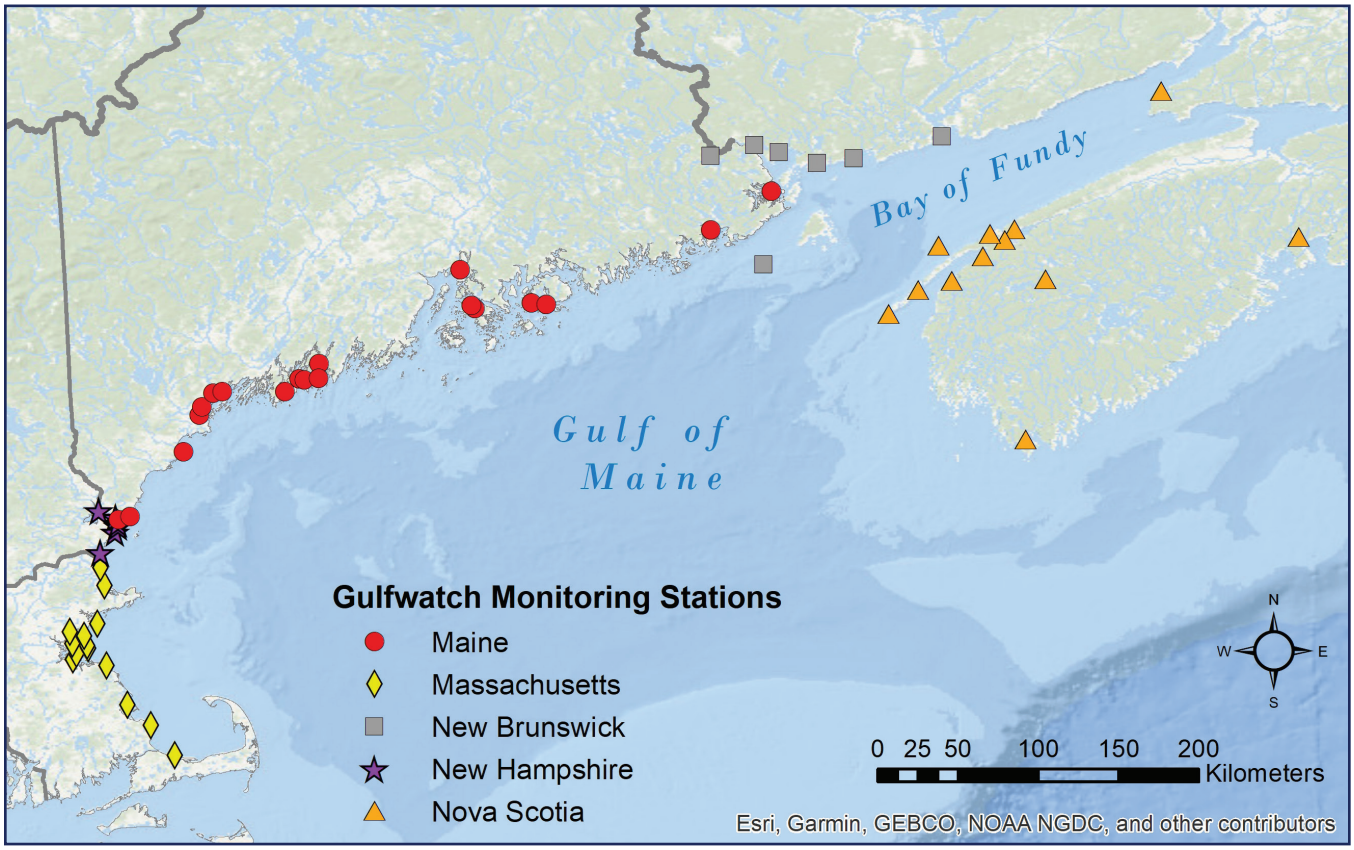


Figure 2. Gulfwatch Program sampling sites: 1993-2016

Introduction

Site BHDI. Credit: NOAA

The Gulf of Maine extends from Cape Sable, Nova Scotia, through New Brunswick, Maine, and New Hampshire to Cape Cod, Massachusetts, and includes the Bay of Fundy and Georges Bank. The immense upwelling of nutrients and the combined productivity of seaweed, salt marsh grasses, and phytoplankton make it one of the world's most productive ecosystems supporting a vast array of organisms, including some of great commercial importance. Commercial fisheries, including aquaculture, as well as tourism and hospitality businesses are principal income-generating enterprises that are important throughout this region and are associated with coastal population growth in the Gulf region. Increases in coastal populations, industrial, and residential development are linked to an increase in municipal wastewater treatment plants contributing to the deteriorating water quality in the Gulf (Pesch and Wells, 2004; Dow and Braasch, 1996; Sowles and Crawford, 1993). Wastewater and industrial effluent discharges, along with septic system releases, and storm water runoff, are some of the major sources of anthropogenic contaminants in aquatic environments in the Gulf of Maine.

Coastal chemical pollution in the Gulf has been assessed and monitored over the years by state, regional and federal organizations for resource and ecosystem management and protection. The National Oceanic and Atmospheric Administration (NOAA) National Status and Trends Program (NS&T) has conducted contaminant assessment and monitoring in the Gulf since 1986 (Kimbrough et al., 2007; Battista et al., 2006), and the US Environmental Protection Agency (EPA) Environmental Monitoring and Assessment Program (EMAP) worked with coastal states to collect field data and conducted ecological monitoring and ecological risk assessment from 1990 to 2006, and in 2010 and 2015. At the state and regional level, the Gulf of Maine Gulfwatch Program established monitoring sites along the Gulf seaboard and conducted the status and trend monitoring of contaminants in blue mussels since 1991 (Jones et al., 2009). These studies have provided relevant data and information to coastal managers and the scientific community, but they were historically focused on legacy contaminants. These legacy pollutants are routinely monitored and regulated and include trace elements ("heavy metals") and persistent organic pollutants such as polychlorinated biphenyls (PCBs), organochlorine pesticides, chlordane, and polycyclic aromatic hydrocarbons (PAHs).

Gulf of Maine. Credit: NOAA

Introduction

As management decisions have helped limit legacy contaminant impacts in coastal waters, it is becoming clear that understanding the hazards of and defining the need to monitor the ever increasing number of new and unregulated contaminants, also known as contaminants of emerging concern (CECs), is critical. Public awareness over the past decade about the environmental fate and health risks (which remain largely unknown) of CECs has only strengthened this need. In addition to lack of adequate scientific techniques to assess their environmental impacts, the sheer number of these chemicals is estimated to be in the tens of thousands (Diamond et al., 2011), compounding the challenges that face researchers and scientific organizations when prioritizing the list of CECs to monitor. Based on EPA recommendations as described in Ankley et al. (2008), classes of CECs to consider for monitoring should include: 1) Persistent organic pollutants (POPs) such as flame retardants, current-use pesticides and industrial by-products, such as perfluorinated and phenolic compounds; 2) Pharmaceuticals and personal care products (PPCPs) such as prescription and/or illegal drugs, sunscreens, and synthetic musks; 3) Veterinary medicines such as antimicrobials, antibiotics, anti-fungals, and growth hormones for animals; 4) Endocrine-disrupting chemicals (EDCs) including synthetic estrogens and androgens as well as many other compounds capable of modulating normal hormonal functions and steroidal synthesis; and 5) Nanoparticles such as carbon nanotubes or nano-scale particulates of which little is known about either their environmental fate or effects. Through a series of pilot studies the MWP has been assessing a suite of CEC compounds in diverse coastal regions for potential consideration for long-term monitoring. This list includes compounds that serve as flame retardants, stain resistant compounds, pharmaceutical and personal care products (PPCPs), endocrine-disrupting chemicals (EDCs), and current-use pesticides (CUPs). The previous MWP CEC studies in Chesapeake Bay and Charleston Harbor (Apeti et al., 2018), the Great Lakes (Kimbrough et al., 2018) and Southern California (Maruya et al., 2016), represent the range of bivalve species and land use types surveyed by the national Mussel Watch Program and indicated a wide range of distribution of CECs in sediment and bivalve shellfish. These results showed that the presence, concentration and distribution of the different classes of CEC compounds are linked to land-use categories in watersheds around the study areas. Although many of the surveyed CECs

Portland, ME. Credit: NOAA

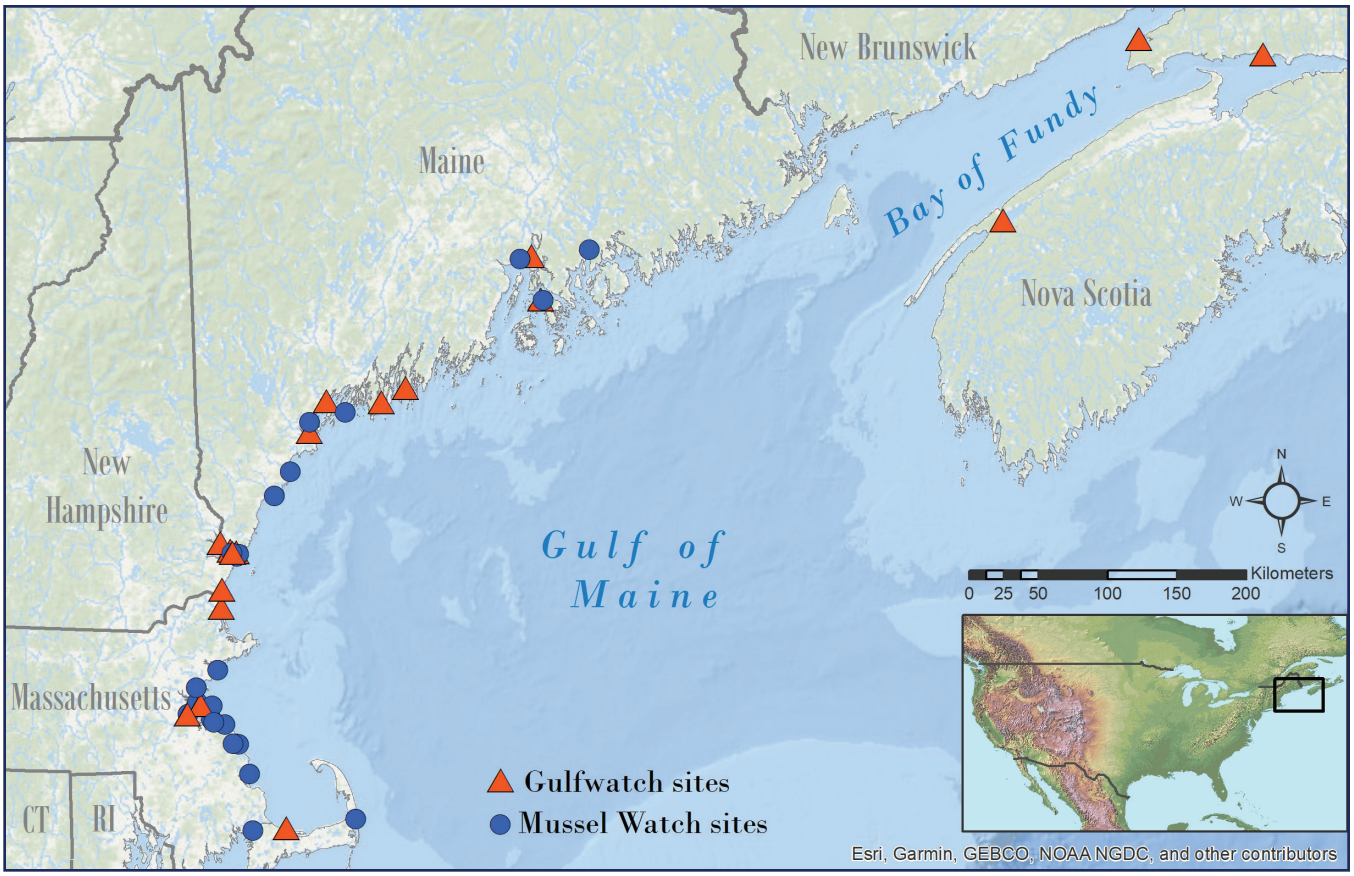


Figure 3. Combined Mussel Watch Program and Gulfwatch Program selected sites for 2015/2016 survey.

were infrequently detected, perhaps as a result of their individual chemical properties, the concentrations of those that were detected increased with urbanization and proximity to storm water discharge outfalls (Apeti et al., 2018; Maruya et al., 2016).

In 2016, the MWP collaborated with the Gulf of Maine Gulfwatch Program to conduct a comprehensive assessment of CECs in the Gulf of Maine (Figure 3). The study was designed within the framework of the MWP regional monitoring approach, which balances short-term flexibility in study design against the cost of broad CEC surveys and combines traditional Mussel Watch sites with those of the Gulfwatch Program. The objectives of study were: 1) assess the presence and distribution of flame retardants, chemicals that enhance stain-resistance, current-use pesticides, PPCPs, and other chemicals associated with human activity that may bioaccumulate in the Gulf of Maine; 2) assess possible links between land-use types and the prevalence and magnitude of CECs in bivalve tissue; 3) conduct inter-jurisdiction comparisons of the CEC results in the Gulf of Maine and weigh the results of this study against previous studies; and 4) make the data electronically available to coastal resource managers in the Gulf of Maine region.

The study leveraged resources from both programs where the Gulfwatch Program provided all the fieldwork and the MWP assumed responsibilty for the analytical analyses and data management. In addition to filling CEC data gaps in the region, this study strengthened federal and state collaboration in monitoring and protecting coastal ecosystems in the Gulf of Maine. Results from this study support the Gulf of Maine Council on the Marine Environment and the Gulfwatch Program's mission to provide "…high quality and relevant data to allow for characterization of the condition of ecosystems in the Gulf of Maine for enhancing marine resource management and protecting public health" (GOMC, 1991).

Methods

Site BHHB. Credit: NOAA

Study Area and Sampling Design

The MWP has 23 historic monitoring sites in the Gulf of Maine, while the Gulfwatch Program has a network of 46 core monitoring sites distributed in the five jurisdictions of Nova Scotia, New Brunswick, Massachusetts, New Hampshire and Maine. The sampling design for this study included the selection of sites from both MWP and Gulfwatch Program. Site selection was conducted in collaboration with resources managers in the region that are part of the Gulf of Maine Council on Marine Environment and it involved a strategic mixture of sites that met both programs' monitoring needs. Sample collection was conducted by the Gulfwatch Program following modified standard protocols utilized by the national MWP and the Gulfwatch Program (Apeti et al., 2012). For this study, 52 blue mussel (*Mytilus* species) samples from 41 different sites were analyzed including 37 composited samples collected in 2016 and 15 frozen composited samples previously collected by the Gulfwatch Program in 2015 (Figure 4, Table 1). The composite sample for each site consisted of approximately 60 mussels collected from three distinct stations (~20 mussels from each station) to create a representative sample. The MWP field activities are designed in a manner so as not to have any significant impact on the environment.

The monitoring sites in both programs were not randomly selected nor designed to target specific pollution sources. The sites were selected in locations with an abundant population of bivalves to allow repetitive sampling and to convey information about the degree of chemical contamination in the general area. However, the spatial distribution of the monitoring sites in diverse waterbodies, tributaries and embayments, sometimes allows the possibility of grouping them into watersheds and conducting land-use assessment. In this study, although sampling design was not based on land-use categories, the presence and distribution of the CECs detected were discussed relative to site proximity to wastewater treatment plants and site location within developed or undeveloped watersheds using land cover measures. Data used for the land-use assessment was obtained from the USGS Multi-Resolution Land Characteristics (MRLC), National Land Cover Database (NLCD) (2011) (https://www.mrlc.gov/nlcd2011.php) raster layer (Homer et al., 2015).

Analytical Methods

In this study, multiple classes of CEC compounds were measured in mussel tissue samples (Table 2). Traditionally, the list of contaminants considered for monitoring would be based on the potential for accumulation, environmental half-life, biodegradation, ecotoxicity and human health information. Since this information does not currently exist or is not fully established for many of the CEC chemicals, NCCOS and the MWP are assessing a list of CECs for which methods are established and for which literature indicates their potential environmental persistence and ecological and human toxicity. During the Southern California Bight project in 2008, a broad scan of diverse classes of CECs (PPCPs, MRES, and phenolic and flame retardant compounds) were evaluated in a variety of matrices (sediment, water, fish and bivalve tissues). This collaborative study provided insight about the detection and concentrations of CECs in different environmental media (Dodder et al., 2014; Maruya et al., 2016), and served as guidance for defining the contaminant lists in these subsequent pilot studies. Detailed descriptions of the analytical methods for CECs measured in this study can be found in Apeti et al. (2018), however, succinct method summaries and background information about each class of CEC are provided further down in this document. Concentrations were blank corrected and any values below the method detection limit (MDL) were qualified as undetected and were assigned a value of zero. The MDL values are determined as specified by EPA Federal Regulations 40 CFR Part 136 (1999). The 99% confidence level MDL is determined based on analysis of a minimum of 7 replicate matrix spikes fortified at 1-10 times the estimated detection limit. The MDL values are "x" times the standard deviation, where "x" is the defined by the student's t-distribution to cover 99% of the distributions of possible values.

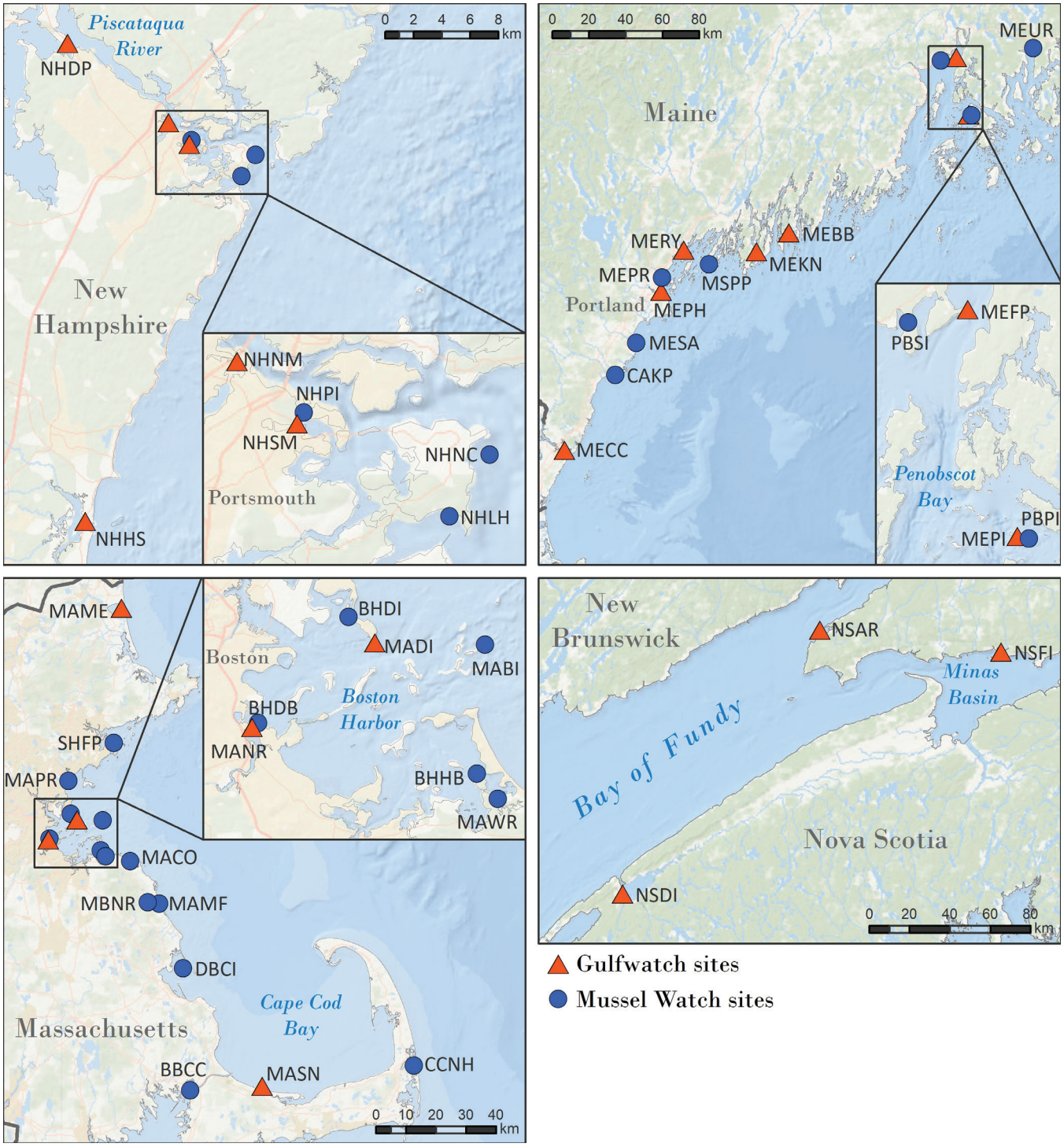


Figure 4. Sites selected in each of the four jurisdictions, New Hampshire, Maine, Massachusetts, and Nova Scotia, for the 2015/2016 survey.

Table 1. Description of MWP (MW) and Gulfwatch (Gw) sites selected for the 2015/2016 survey. MA, Massachusetts; ME, Maine; NH, New Hampshire; NS, Nova Scotia. "●" signifies the site was sampled in that year.

Jurisdiction	Site	General Location	Specific Location	Program	Latitude	Longitude	2015	2016
MA	BBCC	Buzzards Bay	Cape Cod Canal	MW	41.74017	-70.61567		●
MA	BHDB	Boston Harbor	Dorchester Bay	MW	42.30217	-71.03633		●
MA	BHDI	Boston Harbor	Deer Island	MW	42.35733	-70.97300		●
MA	BHHB	Boston Harbor	Hingham Bay	MW	42.27600	-70.88333		●
MA	CCNH	Cape Cod	Nauset Harbor	MW	41.79583	-69.94617		●
MA	DBCI	Duxbury Bay	Clarks Island	MW	42.01367	-70.63650		●
MA	MABI	Boston Harbor	Brewster Island	MW	42.34281	-70.87763		●
MA	MACO	Cohassett	Cohassett	MW	42.25235	-70.79486		●
MA	MADI	Deer Island	Deer Island	Gw	42.34396	-70.95489	●	●
MA	MAME	Merrimack	Merrimack River	Gw	42.81229	-70.82187	●	●
MA	MAMF	Macombers Creek	Macombers Creek	MW	42.15732	-70.70840		●
MA	MANR	Neponset River	Neponset River	Gw	42.30009	-71.04062	●	●
MA	MAPR	Pines River	Pines River	MW	42.43062	-70.98016		●
MA	MASN	Mill Creek	Sandwich	Gw	41.75000	-70.40000	●	
MA	MAWR	Weir River Estuary	Weir River	MW	42.26287	-70.86867		●
MA	MBNR	Massachusetts Bay	North River	MW	42.16033	-70.74250		●
MA	SHFP	Salem Harbor	Folger Point	MW	42.51402	-70.84416		●
ME	CAKP	Cape Arundel	Kennebunkport	MW	43.34533	-70.47433		●
ME	MEBB	Boothbay Harbor	Boothbay Harbor	Gw	43.85130	-69.62688	●	●
ME	MECC	Piscataqua / Salmon Falls	Clark Cove	Gw	43.07700	-70.72389	●	●
ME	MEFP	Penobscot	Fort Point	Gw	44.46814	-68.80958	●	
ME	MEKN	Kennebec	Perkins Island	Gw	43.78483	-69.78492	●	●
ME	MEPH	Stroudwater-Fore	Portland Harbor	Gw	43.64438	-70.25153	●	●
ME	MEPI	Penobscot	Pickering Island	Gw	44.26699	-68.74841	●	
ME	MEPR	Presumpscot	Presumpscot River	MW	43.69155	-70.24677		●
ME	MERY	Royal River	Royal River	Gw	43.79000	-70.14121	●	
ME	MESA	Saco	Saco River	MW	43.45990	-70.37257		●
ME	MEUR	Union River	Union River	MW	44.50013	-68.43204		●
ME	MSPP	Merriconeag Sound	Potts Point	MW	43.73955	-70.01678		●
ME	PBPI	Penobscot Bay	Pickering Island	MW	44.26483	-68.73367		●
ME	PBSI	Penobscot Bay	Sears Island	MW	44.45667	-68.88317		●
NH	NHDP	Piscataqua River	Dover Point	Gw	43.11966	-70.82738	●	●
NH	NHHS	Hampton-Seabrook Estuary	Hampton-Seabrook Estuary	Gw	42.89725	-70.81624	●	●
NH	NHLH	Piscataqua River	Little Harbor	MW	43.05798	-70.71697		●
NH	NHNC	New Castle	New Castle	MW	43.06786	-70.70817		●
NH	NHNM	North Mill Pond	North Mill Pond	Gw	43.08283	-70.76345	●	
NH	NHPI	Pierce island	Pierce island	MW	43.07465	-70.74881		●
NH	NHSM	South Mill Pond	South Mill Pond	Gw	43.07283	-70.75028	●	
NS	NSAR	Chignecto Bay	Apple River	Gw	45.46667	-64.87222		●
NS	NSDI	Annapolis Basin	Digby	Gw	44.63333	-65.75000		●
NS	NSFI	Minas/Cobequid Shore	Five Islands	Gw	45.39750	-64.06600		●

Table 2. Contaminants of emerging concern measured as part of the Mussel Watch Program pilot studies.

Compound class	Compounds
Alkylphenol Compounds (APs)	4-NP, 4-n-OP, NP1EO, NP2EO
Alternative Flame Retardants (AFRs)	alpha-HBCD, beta-HBCD, gamma-HBCD, BTBPE, TBB, TBPH, TCEP, TCPP, TDCPP
Current-Use Pesticides (CUPs)	Ametryn, Atrazine, Azinphos-Methyl, Captan, Chlorothalonil, Chlorpyrifos, Chlorpyrifos-Methyl, Chlorpyrifos-Oxon, Cyanazine, Cypermethrin, Dacthal, Desethylatrazine, Diazinon, Diazinon-Oxon, Dimethoate, Disulfoton, Disulfoton Sulfone, Ethion, Fenitrothion, Fonofos, Hexazinone, Malathion, Methoxychlor, Metribuzin, Parathion-Ethyl, Parathion-Methyl, Permethrin, Perthane, Phosmet, Pirimiphos-Methyl, Quintozene, Simazine, Tecnazene
Per- and Polyfluoroalkyl Substances (PFASs)	PFBS, PFDA, PFDODA, PFDS, PFHPA, PFHXA, PFHXS, PFNA, PFOA, PFOS, PFOSA, PFUNDA
Pharmaceuticals and Personal Care Products (PPCPs)	10-hydroxy-amitriptyline, 17α-DihydroEquilin, 17α-estradiol, 17α-Ethynyl estradiol, 17β-estradiol, 2-Hydroxy-ibuprofen, Acetaminophen, Albuterol, Allyl Trenbolone, Alprazolam, Amitriptyline, Amlodipine, Amphetamine, Androstenedione, Androsterone, Atenolol, Atorvastatin, Azithromycin, Benzoylecgonine, Benztropine, Betamethasone, Bisphenol-A, Busulfan, Caffeine, Carbadox, Carbamazepine, Cimetidine, Ciprofloxacin, Citalopram, Clarithromycin, Clinafloxacin, Clonidine, Clotrimazole, Cloxacillin, Cocaine, Codeine, Cotinine, DEET, Dehydronifedipine, Desogestrel, Diazepam, Diethylstilbestrol, Digoxigenin, Digoxin, Diltiazem, Diphenhydramine, Enalapril, Enrofloxacin, Equilenin, Equilin, Erythromycin, Estriol, Estrone, Etoposide, Flumequine, Fluocinonide, Fluoxetine, Fluticasone propionate, Furosemide, Gemfibrozil, Glipizide, Glyburide, Hydrochlorothiazide, Hydrocodone, Hydrocortisone, Ibuprofen, Lomefloxacin, Meprobamate, Mestranol, Metformin, Methylprednisolone, Metprolol, Miconazole, N-Desmethyldiltiazem, Naproxen, Norfloxacin, Norfluoxetine, Norgestimate, Norgestrel, Norverapamil, Ofloxacin, Ormetoprim, Oxacillin, Oxolinic Acid, Oxycodone, Paraxanthine, Paroxetine, Penicillin G, Penicillin V, Prednisolone, Prednisone, Progesterone, Promethazine, Propoxyphene, Propranolol, Ranitidine, Roxithromycin, Sarafloxacin, Sertraline, Simvastatin, Sulfachloropyridazine, Sulfadiazine, Sulfadimethoxine, Sulfamerazine, Sulfamethazine, Sulfamethizole, Sulfamethoxazole, Sulfanilamide, Sulfathiazole, Testosterone, Theophylline, Thiabendazole, Triamterene, Triclocarban, Triclosan, Trimethoprim, Tylosin, Valsartan, Venlafaxine, Verapamil, Warfarin
Polybrominated Flame Retardants (BFRs)	Polybrominated Biphenyls (PBBs): PBB 1, PBB 2, PBB 3, PBB 4, PBB 7, PBB 9, PBB 10, PBB 15, PBB 18, PBB 26, PBB 30, PBB 31, PBB 49, PBB 52, PBB 53, PBB 77, PBB 80, PBB 103, PBB 155 Polybrominated Diphenyl Ethers (PBDEs): PBDE-1, PBDE-2, PBDE-3, PBDE-7, PBDE-8, PBDE-10, PBDE-11, PBDE-12, PBDE-13, PBDE-15, PBDE-17, PBDE-25, PBDE-28, PBDE-30, PBDE-32, PBDE-33, PBDE-35, PBDE-37, PBDE-47, PBDE-66, PBDE-71/49, PBDE-75, PBDE-77, PBDE-85, PBDE-99, PBDE-100, PBDE-116, PBDE-118, PBDE-119, PBDE-126, PBDE-138, PBDE-153, PBDE-154, PBDE-155, PBDE-166, PBDE-181, PBDE-183, PBDE-190, PBDE-194, PBDE-195, PBDE-196, PBDE-197, PBDE-198/199/203/200, PBDE-201, PBDE-202, PBDE-204, PBDE-205, PBDE-206, PBDE-207, PBDE-208, PBDE-209

Data Analysis

Data management and analysis were conducted using a combination of R version 3.4.4 (R Core Team, 2013), Microsoft Excel (2016) and ArcGIS (ESRI, 2011). For the few sites that were sampled in both 2015 and 2016, if both sample concentrations were above the MDL then the site mean of both years was used for analysis. If the sample concentration for only one of the years was above the MDL, the single concentration value above MDL was used for analysis.

To analyze any relationships between land-use and chemical concentrations, percent impervious surface and land-use values were determined for each site. Land-use classification procedures used in this study are detailed in Edwards et al. (2014) and Edwards et al. (2016). Briefly, land-use and percent impervious surface was assigned by clipping the United States Geological Survey (USGS) Multi-Resolution Land Characteristics Consortium (MRLC) National Land Cover Database (NLCD) (2011) (MRLC; https://www.mrlc.gov/) by a 1, 2, 3, 4, and 5 km buffer around each site. Impervious surface was determined by calculating the average percent impervious surface of the 30-meter pixels within each buffer. Land-use estimates were recalculated from each land-use and land cover class contained within each clipped buffer. The land-use classes for each monitoring site were re-classified and aggregated into five distinct land-use categories (agriculture, low developed, undeveloped and urban and open-water) following the categories developed from the Anderson Level I land cover and land-use classification scheme (Anderson et al., 1976) (Table 3). Finally, using a Principle Components Analysis (PCA) based on the five land-use category percentages, each site was assigned a mutually exclusive land-use category for each radius sized buffer (Table 4). Since the Anderson land-use category combinations of undeveloped/agricultural and low-developed/urban were closely correlated in the PCA results, the final land-use designations were simplified to undeveloped (undeveloped and agricultural), developed (low-developed and urban) and open-water. Additionally, the Environmental Protection Agency (EPA) Facility Registry Service (FRS) Wastewater Treatment Plants (WWTP) data layer (https://catalog.data.gov/dataset/epa-facility-registry-service-frs-wastewater-treatment-plants) was used to evaluate the proximity of WWTPs and combined sewer outfalls (CSO) to the monitoring sites. Discharges from WWTPs are considered as the most important sources of CECs pollutants into the aquatic environment. Unfortunately, none of the aforementioned data layers contained information for Nova Scotia.

When at least 12 (~30%) of the sites had detects, total and compound class detection frequencies and individual compound concentrations were tested for normality using Shapiro-Wilk tests ($p<0.05$). The effects of land-use on normally distributed data was analyzed using linear regressions ($p<0.05$) and one-way ANOVAs ($p<0.05$). If the assumptions of the parametric statistics were not met, a non-parametric Spearman's rank correlation ($p<0.05$) and a Kruskall-Wallis one-way analysis of variance ($p<0.05$) were applied.

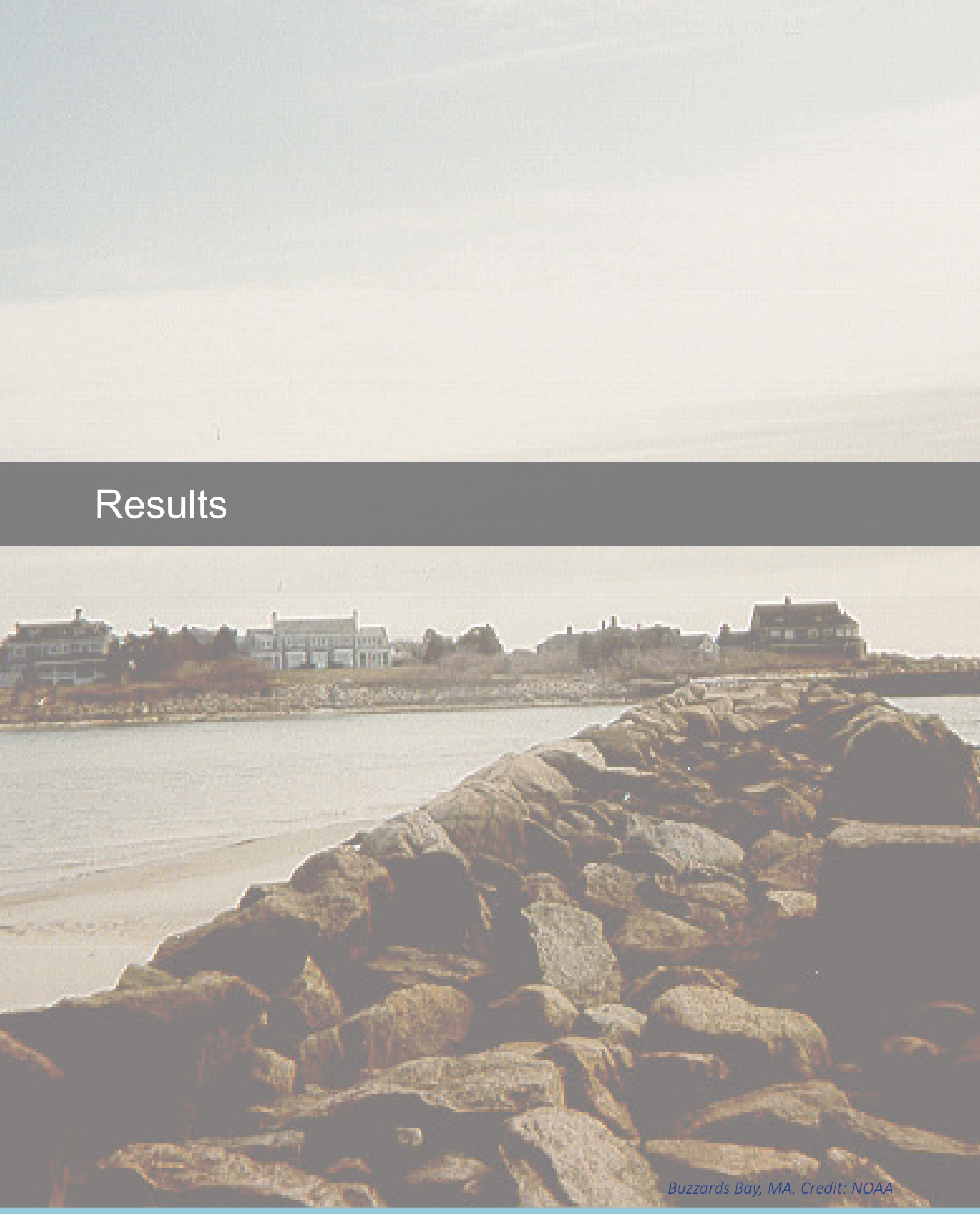
Table 3. Study Sites Land-use and Land cover Classification Scheme (Anderson et al., 1976).

Land-use categories	NLCD 2011 Land-use and land cover classification groups
Urban	23 - Developed Medium Intensity; 24 - Developed High Intensity
Low-developed	21 - Developed, Open Space; 22 - Developed, Low Intensity
Agriculture	81 - Pasture/Hay; 82 - Cultivated Crops
Undeveloped	31 - Barren Land; 41 - Deciduous Forest; 42 - Evergreen Forest; 43 - Mixed Forest; 52 - Shrub/Scrub; 71 - Herbaceous/Grassland; 90 - Woody Wetlands; 95 - Emergent Herbaceous Wetlands
Open-water	11 - Open-water

Table 4. Percent impervious surface and land-use classification of monitoring sites based on the reclassification techniques used in this study using 1, 2, 3, 4 and 5 km buffers. Sites classified as NA (not applicable) were outside of the continental U.S. and beyond the limits of the data layers. MA, Massachusetts; ME, Maine; NH, New Hampshire; NS, Nova Scotia; D, Developed; U, Undeveloped; O, Open-water

Jurisdiction	Site	Percent Impervious Surface					Land-use Classification				
		1 Km	2 Km	3 Km	4 Km	5 Km	1 Km	2 Km	3 Km	4 Km	5 Km
MA	BBCC	34	24	21	17	14	D	D	D	D	U
MA	BHDB	17	34	37	39	39	O	D	D	D	D
MA	BHDI	11	14	15	16	17	O	O	O	O	O
MA	BHHB	9	12	11	11	10	O	O	O	O	O
MA	CCNH	13	9	7	7	7	U	U	U	U	U
MA	DBCI	0	2	2	2	2	O	O	O	O	O
MA	MABI	0	0	0	0	1	O	O	O	O	O
MA	MACO	7	8	8	7	6	U	D	D	U	U
MA	MADI	12	5	4	4	6	O	O	O	O	O
MA	MAME	11	5	4	7	7	O	O	O	U	U
MA	MAMF	4	3	5	5	5	O	O	O	U	U
MA	MANR	27	41	42	42	41	D	D	D	D	D
MA	MAPR	13	12	23	32	34	U	O	O	D	D
MA	MASN	0	3	3	3	3	O	O	O	U	U
MA	MAWR	9	13	13	12	11	U	D	D	D	D
MA	MBNR	4	4	6	6	6	U	U	U	U	U
MA	SHFP	14	15	12	12	15	D	O	O	O	O
ME	CAKP	6	5	4	3	2	U	U	U	U	U
ME	MEBB	15	7	4	3	2	U	U	U	U	U
ME	MECC	17	14	11	12	11	D	D	U	D	U
ME	MEFP	0	0	1	1	1	O	O	O	U	U
ME	MEKN	0	0	0	1	1	O	O	U	U	U
ME	MEPH	33	41	34	25	22	D	D	D	D	D
ME	MEPI	0	0	0	0	0	O	O	O	O	O
ME	MEPR	12	12	12	16	17	D	D	D	D	D
ME	MERY	0	1	3	4	4	U	O	U	U	U
ME	MESA	5	5	3	3	2	O	O	O	U	U
ME	MEUR	2	2	2	3	4	U	U	U	U	U
ME	MSPP	2	3	2	1	1	O	O	O	O	O
ME	PBPI	0	0	0	0	0	O	O	O	O	O
ME	PBSI	2	6	4	3	3	O	O	U	U	U
NH	NHDP	9	7	7	9	10	U	U	U	U	U
NH	NHHS	19	11	7	8	10	O	O	O	U	U
NH	NHLH	7	4	5	7	8	U	O	O	U	U
NH	NHNC	3	4	5	6	7	O	O	O	U	U
NH	NHNM	39	34	29	24	21	D	D	D	D	D
NH	NHPI	29	27	24	19	17	D	D	D	D	D
NH	NHSM	27	28	24	20	17	D	D	D	D	D
NS	NSAR	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
NS	NSDI	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA
NS	NSFI	NA	NA	NA	NA	NA	NA	NA	NA	NA	NA

Results



Buzzards Bay, MA. Credit: NOAA

Alkylphenol Compounds (APs)

Chemical Description

Alkylphenols are a class of chemicals used in detergents and surfactants in industrial processes. Some household detergents (i.e. laundry soaps) also include APs. The most common sources of APs to aquatic systems are wastewater and septic system discharges (Ying et al., 2002). These compounds tend to be persistent in the environment, have a strong affinity for suspended particles, and are well preserved in bottom sediments (Ying et al., 2002). In the environment, alkylphenol ethoxylate surfactants biodegrade into more environmentally stable metabolites, such as the alkylphenol n-ethoxylates, alkylphenoxy acetic and alkylphenoxy-polyethoxy acetic acids, and alkylphenols (EPA, 2014a). This study focused on four AP metabolites in mussel tissues (Table 5). Two of the compounds 4-nonylphenol (4-NP) and 4-n-octylphenol (4-n-OP) are degradation products of 4-nonylphenol mono-ethoxylate (NP1E0) and 4-nonylphenol di-ethoxylate (NP2E0), which are byproducts of the parent alkylphenol polyethoxylate. These degradation products are reported to be more toxic than the parent compounds and act as hormone mimics (Ying et al., 2002). Alkylphenols are shown to have estrogenic endocrine-disrupting effects on vertebrate organisms, and they have been linked to severe decreases in lobster larval survival and juvenile lobster hormonal changes (Laufer et al., 2013).

Among the diverse group of alkylphenols, nonylphenol ethoxylates (NPEOs), metabolites of commercial detergents, and their environmental degradation products nonylphenols (NPs), were included in the EPA New Use Rules list of 15 toxic AP compounds (EPA, 2014a). In this study, the MWP measured two NPEO and two NP compounds (Table 3) for which analytical methods are well established. The analyses were conducted by the NCCOS' chemistry laboratory in Charleston, SC based on published methods by Petrovic et al. (2002) and Loyo-Rosales et al. (2003).

Table 5. AP compounds tested.

Chemical code	Chemical name
4-n-OP	4-n-octylphenol
4-NP	4-nonylphenol
NP1EO	4-nonylphenol mono-ethoxylate
NP2EO	4-nonylphenol di-ethoxylate

Presence and distribution of APs in mussel tissue: GULF-WIDE ASSESSMENT

Table 6. AP compounds Gulf-wide frequency of detection in mussel tissue.

Compound	Number of Detects	Number of Sampled Sites	Frequency (%)
NP1EO	13	40	32.5
NP2EO	2	40	5
4-n-OP	1	40	2.5
Compound Class Total	16	160	10

Number of compounds detected:
3/4

Number of sites with detects:
13/40

Most detected compound:
NP1EO

Table 7. AP compounds number of detects in mussel tissue at each site.

Site	State	Number of Detects	Number of Compounds Analyzed
NHHS	NH	2	4
NHSM	NH	2	4
MEPR	ME	2	4
MANR	MA	1	4
BHDI	MA	1	4
MAME	MA	1	4
NHNC	NH	1	4
NHNM	NH	1	4
NHDP	NH	1	4
MECC	ME	1	4
CAKP	ME	1	4
MEPH	ME	1	4
MEKN	ME	1	4

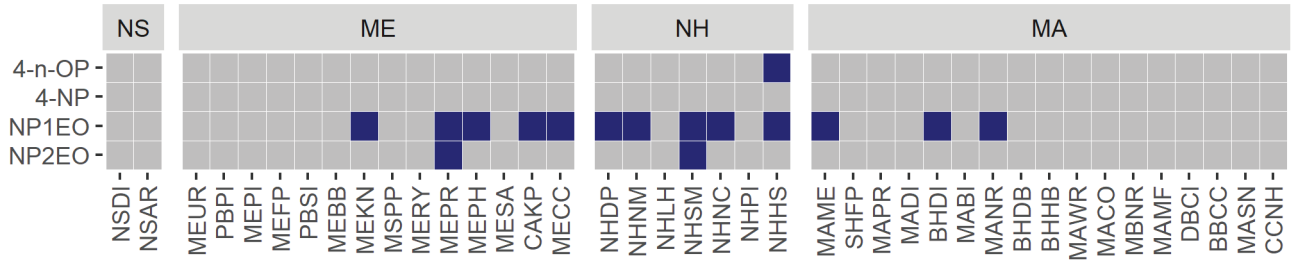


Figure 5. Distribution map showing presence (■) and absence (■) of AP compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

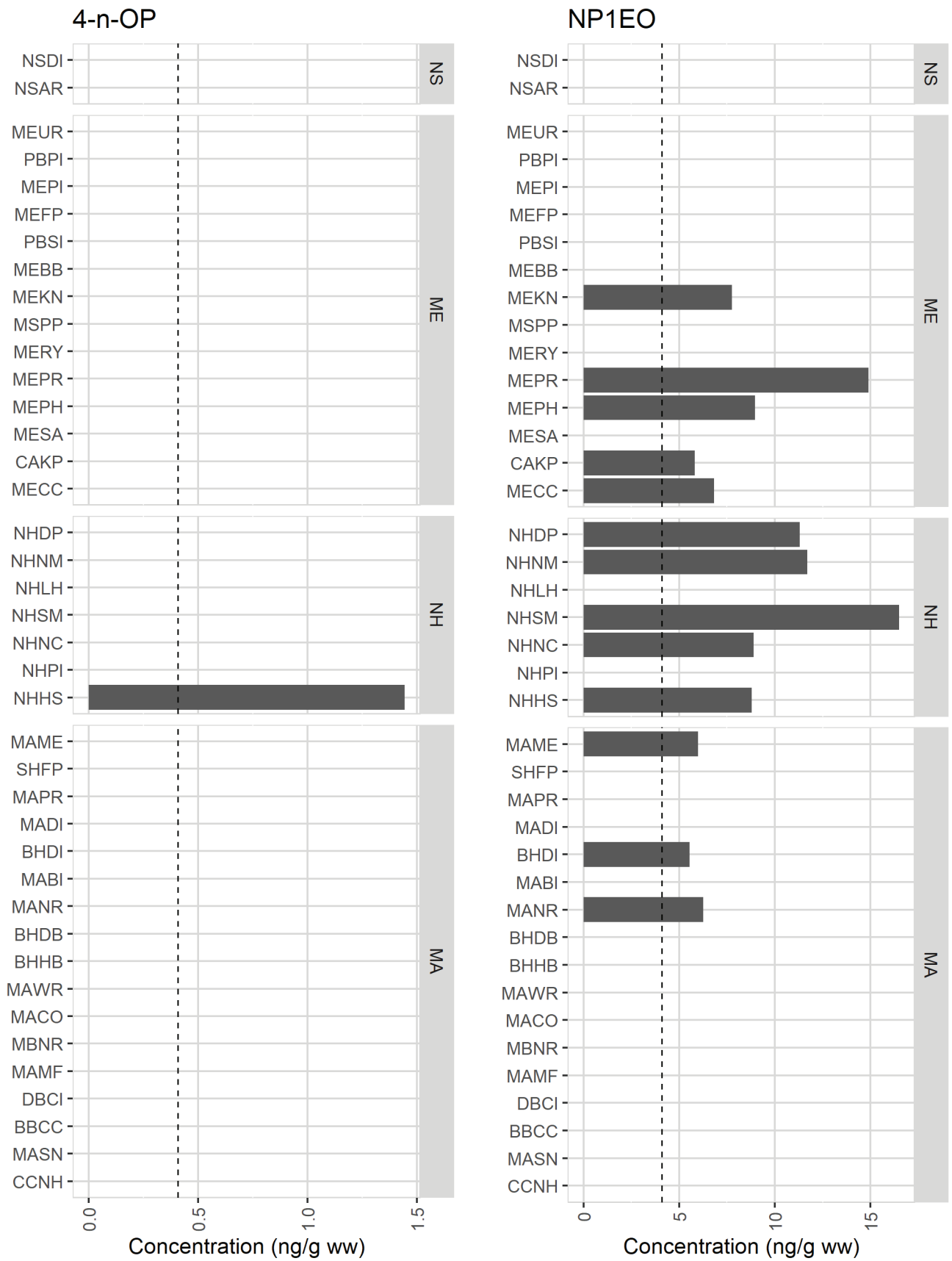
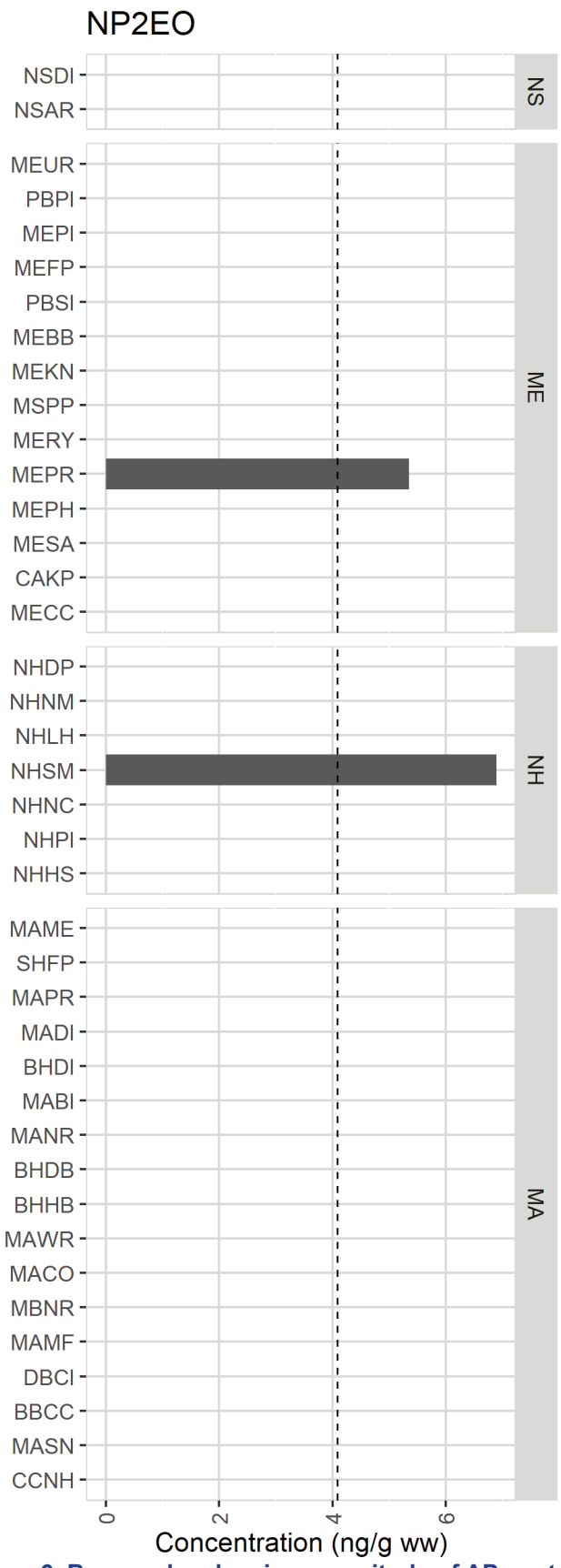


Figure 6. Bar graphs showing magnitude of AP contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

Gulf-wide Assessment



Summary of APs in mussel tissue

- Due to insufficient sample mass at site NSFI, AP contaminants were measured in 40 mussel tissue samples (17 in MA, 14 in ME, 7 in NH and 2 in NS).
- Three of the four AP contaminants, 4-nonylphenol mono-ethoxylate (NP1E0), 4-nonylphenol di-ethoxylate (NP2E0), and 4-n-octylphenol (4-n-OP) were found at above detection limits in mussel from different locations across the Gulf of Maine (Table 6, Figure 5).
- 4-nonylphenol mono-ethoxylate (4-NP1EO) was the most frequently detected of the AP contaminants with an estimated 32.5% frequency of detection Gulf-wide (Table 6). 4-NP1EO was found at a total of 13 monitoring sites across MA, NH and ME (Table 7).
- 4-nonylphenol di-ethoxylate was detected at two sites, the MEPR site in ME, and the NHSM site in NH (Figure 5).
- 4-n-octylphenol was only detected at site NHHS in NH (Figure 5).
- 4-nonylphenol, a degradation product of 4-nonylphenol mono-ethoxylate, was the only AP compound that was not detected in mussel tissue from the Gulf of Maine.
- The magnitude of AP contaminants measured varied in mussel tissue across the Gulf (Figure 6). A maximum concentration of 16.5 ng/g ww was recorded at the South Mill Pond (NHSM) site in NH for 4-NP1EO (Figure 6, Appendix 1).
- AP detection frequencies and NP1EO concentrations were significantly positively correlated with percent impervious surface for every buffer size (p= .01 - .031), however correlations were weak (rho = 0.34 - 0.41) (Appendix 6).
- Sites with detected AP compounds were located in developed, undeveloped and open-water land-use categories (Table 4), however, AP detection frequencies and NP1EO concentrations were higher at developed land-use sites than open-water or undeveloped sites in a 1 km buffer (p=.043, p=.022) (Appendix 6). Both NP2EO detections were at sites associated with the developed land-use category.

Figure 6. Bar graphs showing magnitude of AP contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

Jurisdiction-specific assessments: MASSACHUSETTS SUMMARY

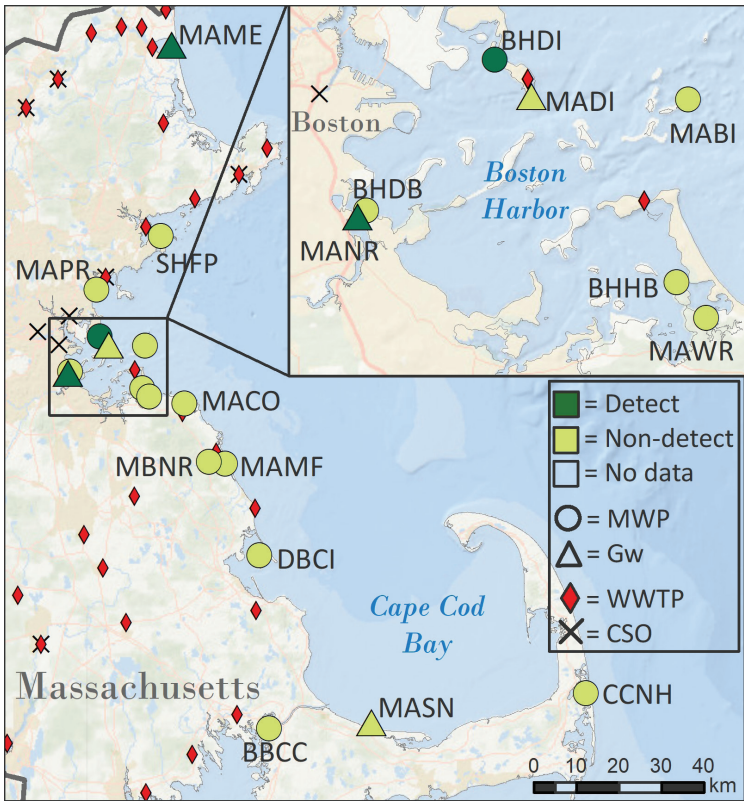


Figure 7. Map of MA jurisdiction highlighting location of sites with AP detection in mussel tissue.

Table 8. AP compounds frequency of detection in mussel tissue from MA jurisdiction (n = 17).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
NP1EO	3	17	17.6

Summary of APs in Massachusetts

- A total of 17 mussel tissue samples were tested for AP contaminants in MA, and only three sites recorded detected levels of AP (Figure 7).
- Among the four APs compounds tested, 4-nonylphenol mono-ethoxylate (NP1EO) was the only one detected (Table 8).
- In MA, 4-nonylphenol mono-ethoxylate was detected in mussels from the Neponset River (MANR), the Boston Harbor Deer Island (BHDI), and the Merimac River (MAME) sites (Figure 7).
- With only three of the 17 samples showing detectable levels of APs, detection frequency of 17.6% (Table 8) indicated that the phenolic compounds tested were sparsely distributed in the coastal water of MA.
- The magnitude of 4-nonylphenol mono-ethoxylate detected was similar, varying from 5.55 ng/g ww in BHDI and 6.25 ng/g ww in MANR (Figure 6, Appendix 1).
- MANR was associated with the developed land-use category (Table 4). Sites BHDI and MAME were associated with open-water or undeveloped land-use categories, however, BHDI is located in Boston Harbor and MAME is located at the mouth of the Merrimack River which recieves discharge from multiple wastewater treatment plants (Figure 7).

Jurisdiction-specific assessments: NEW HAMPSHIRE SUMMARY

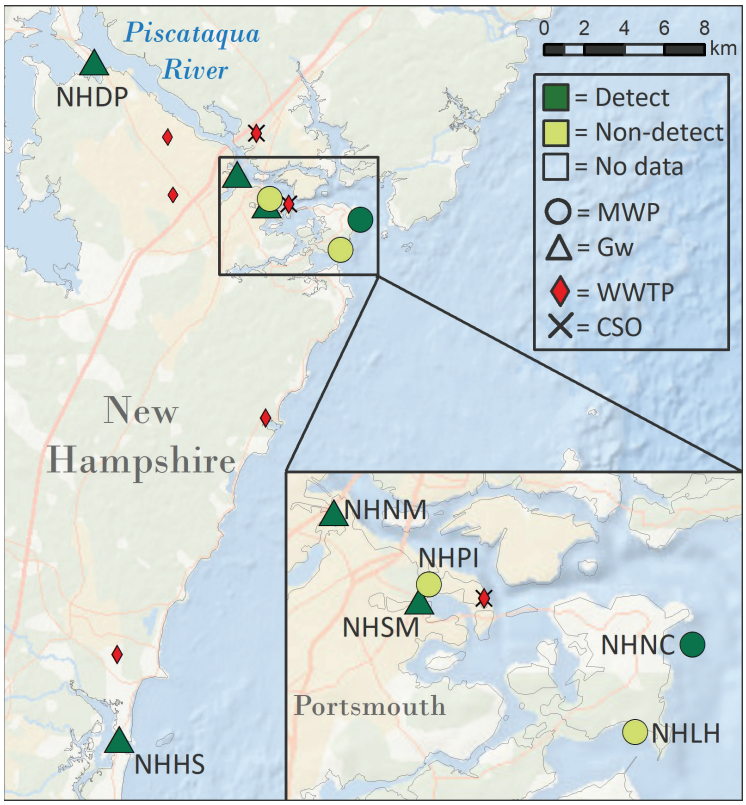


Figure 8. Map of NH jurisdiction highlighting location of sites with AP detection in mussel tissue.

Table 9. AP compounds frequency of detection in mussel tissue from NH jurisdiction (n = 7).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
NP1EO	5	7	71.4
4-n-OP	1	7	14.3
NP2EO	1	7	14.3

Summary of APs in New Hampshire

- A total of 7 mussel tissue samples were tested for AP contaminants in NH, and samples from five sites were positively tested for at least one AP compounds (Figure 8).
- Three of the four AP contaminants, 4-nonylphenol mono-ethoxylate (NP1E0), 4-nonylphenol di-ethoxylate (NP2E0), and 4-n-octylphenol (4-n-OP) were detected in NH (Table 9).
- The 4-nonylphenol di-ethoxylate (NP2E0) and 4-n-octylphenol (4-n-OP) were detected each at a single site (Table 9). 4-nonylphenol di-ethoxylate was detected at South Mill Pond (NHSM) at a concentration of 6.88 ng/g ww, while 4-n-octylphenol was detected at the Hampton-Seabrook Estuary site (NHHS) at 1.44 ng/g ww (Appendix 1).
- Detected at Hampton-Seabrook Estuary (NHHS), New Castle (NHNC), South Mill Pond (NHSM), North Mill Pond (NHNM) and Piscataqua River (NHDP) sites, 4-nonylphenol mono-ethoxylate (NP1E0) was the most detected AP compound in NH with a detection frequency of 71.4% (Table 9). The magnitude of the detected NP1E0 varied from 8.80 ng/g ww at NHHS site to a maximum concentration of 16.50 ng/g ww at NHSM (Figure 6, Appendix 1).
- In NH, NHNM and NHSM were associated with the developed land-use category (Table 4). Additionally, both sites are in close proximity to or downstream from wastewater treatment plants (Figure 8). NHHS, NHNC, NHDP were considered undeveloped and open-water.

Jurisdiction-specific assessments: MAINE SUMMARY

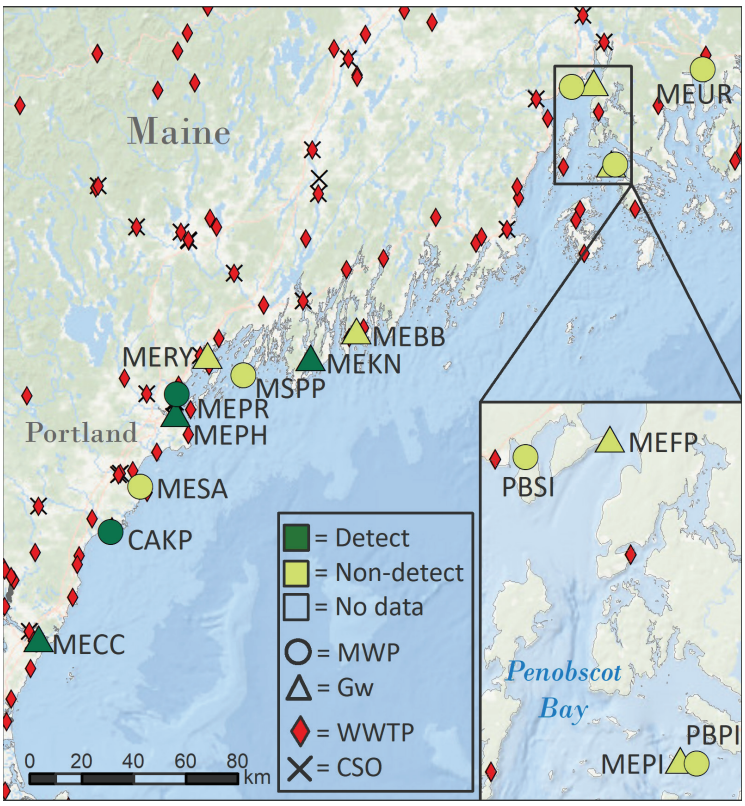


Figure 9. Map of ME jurisdiction highlighting location of sites with AP detection in mussel tissue.

Table 10. AP compounds frequency of detection in mussel tissue from ME jurisdiction (n = 14).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
NP1EO	5	14	35.7
NP2EO	1	14	7.1

Summary of APs in Maine

- In ME, a total of 14 mussel tissue samples were tested for AP contaminants, and samples from five sites were tested positively for at least one AP compounds (Figure 9).
- Of the four AP compounds tested, only two 4-nonylphenol mono-ethoxylate (NP1E0) and 4-nonylphenol di-ethoxylate (NP2E0) were detected in mussels from NH (Table 10).
- Detected at the Presumpscot River (MEPR), Kennebec Prekins Island (MEKN), Stroudwater-Fore Portland Harbor (MEPH), Cape Arundel Kennebunkport (CAKP) and Clark Cove (MECC) sites, 4-nonylphenol mono-ethoxylate (NP1E0) was the most detected AP compound in ME with the frequency of detection of 35.7% (Table 10). The magnitude of the detected NP1E0 varied from 5.81 ng/g ww at CAKP site to a maximum concentration of 14.9 ng/g ww at MEPR (Figure 6, Appendix 1).
- The 4 nonylphenol di-ethoxylate (NP2E0) was only detected at the Presumpscot River (MEPR) site at a concentration of 5.35 ng/g ww (Figure 9, Appendix 1).
- MEPR, MEPH, MECC were primarily associated with the developed land-use category (Table 4). CAKP and MEKN were considered undeveloped or open-water, however, dischargers from wastewater treatment plants and outfalls located in the watershed (Figure 9) may have influenced these sites.

Jurisdiction-specific assessments: NOVA SCOTIA SUMMARY

Figure 10. Map of NS jurisdiction highlighting location of sites with AP detection in mussel tissue.

Summary of APs in Nova Scotia

- Mussel tissue samples from two monitoring sites in NB/NS were measured for AP contaminants.
- None of the four AP contaminants tested were detected in the jurisdictions of NS (Figure 10).

Site CAKP. Credit: NOAA

Alternative Flame Retardants (AFRs)

Chemical Description

Alternative flame retardants are added to a wide variety of industrial and consumer products, such as textiles, rugs, furniture and plastics (de Wit, 2002). For this study, several groups of chemicals were combined under the title of alternative flame retardants, including hexabromocyclododecanes (HBCDs) and chlorinated organophosphate (CPP) chemicals (Table 11). Hexabromocyclododecanes (HBCDs) are primarily used in household consumer products such as upholstery, polystyrene, and textiles. HBCDs are ubiquitous in the environment, but their ecotoxicity is not well understood. The chlorinated organophosphate flame retardants such as tris(1,3-dichloroisopropyl)phosphate (TDCPP) are mainly used as additives in textiles. As additives, chlorinated organophosphate flame retardants tend to leach out over time into water and air. In the environment, TDCPP can accumulate in animal fat tissues (Andresen et al. 2004). The brominated flame retardants 2-ethylhexyl tetrabromobenzoate (TBB) and 2-ethylhexyl 3,4,5,6-tetrabromophthalate (TBPH) and their metabolites have anti-androgenic and anti-thyroid hormonal activities properties (Klopcic et al., 2016). The chemicals TBB and TBPH were introduced as replacements for the PBDEs and functionally reduce flammability in products like electronic devices, textiles, plastics, coatings and polyurethane foams.

AFR analyses were performed by TDI-Brooks International Inc. following procedures used by the NOAA NS&T Program (Kimbrough et al., 2007).

Table 11. AFR compounds tested.

Chemical code	Chemical name
alpha-HBCD	α-Hexabromocyclododecane
beta-HBCD	β-Hexabromocyclododecane
gamma-HBCD	γ-Hexabromocyclododecane
BTBPE	1,2-Bis(2,4,6-tribromophenoxy)ethane
TBB	4,5,6,7-tetrabromobenzotriazole
TBPH	bis(2-ethylhexyl) tetrabromophthalate
TCEP	Tris(2-chloroethyl) phosphate
TCPP	Tris (chloroisopropyl) phosphate
TDCPP	Tris(1,3-dichloroisopropyl)phosphate

Presence and distribution of AFRs in mussel tissue: GULF-WIDE ASSESSMENT

Table 12. AFR compounds Gulf-wide frequency of detection in mussel tissue.

Compound	Number of Detects	Number of Sampled Sites	Frequency (%)
TBB	6	38	15.8
TBPH	1	38	2.6
Compound Class Total	7	342	2

Table 13. AFR compounds number of detects in mussel tissue at each site.

Site	State	Number of Detects	Number of Compounds Analyzed
MEKN	ME	2	9
DBCI	MA	1	9
BHHB	MA	1	9
BHDI	MA	1	9
MADI	MA	1	9
MEPR	ME	1	9

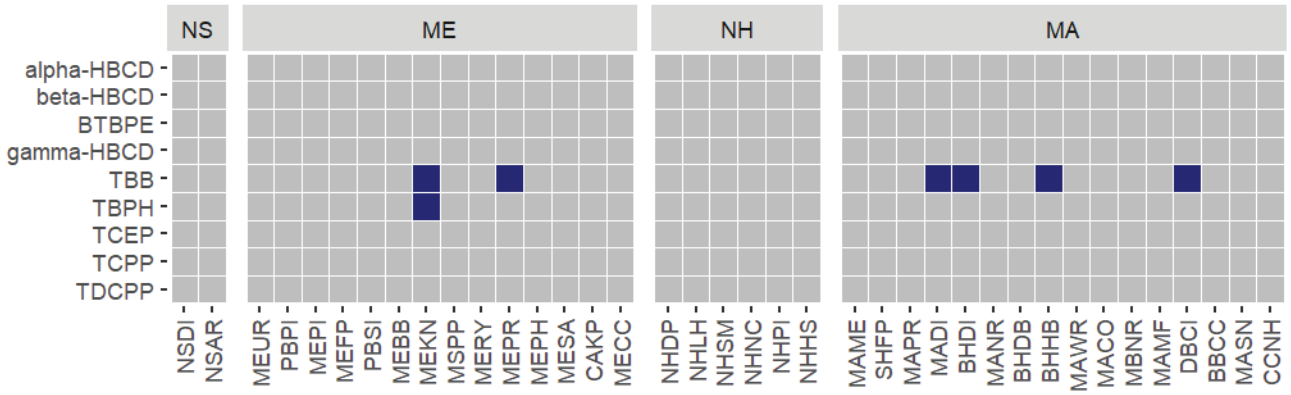


Figure 11. Distribution map showing presence (■) and absence (■) of AFR compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

Number of compounds detected:
2/9

Number of sites with detects:
6/38

Most detected compound:
TBB

GULF-WIDE ASSESSMENT

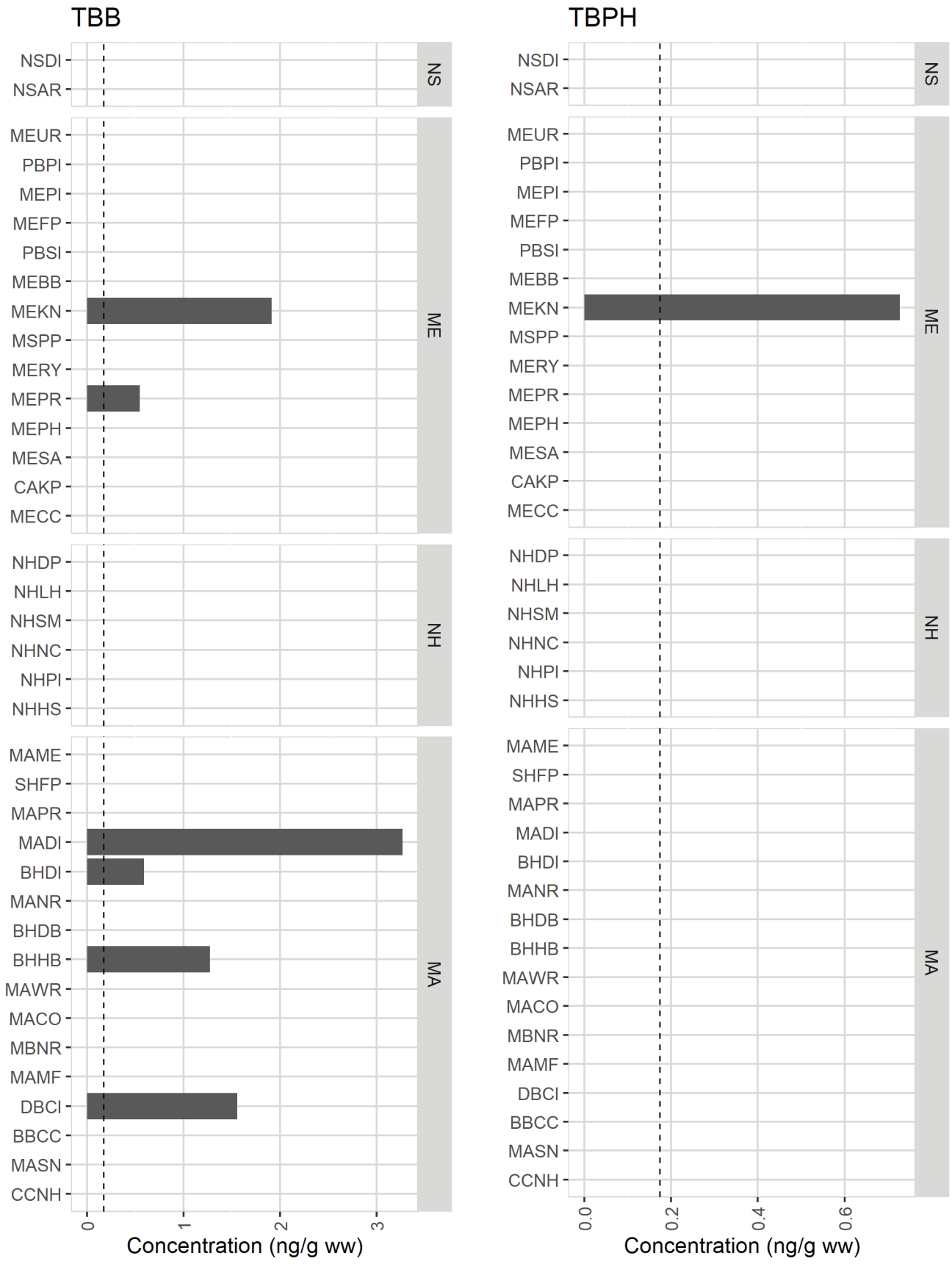


Figure 12. Bar graphs showing magnitude of AFR contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

Summary of AFRs in mussel tissue

- Due to insufficient sample mass at MABI, NHNM and NSFI, AFR contaminants were measured in 38 mussel tissues (16 in MA, 14 in ME, 6 in NH and 2 in NS).
- Of the nine AFR contaminants only two, 2-ethylhexyl tetrabromobenzoate (TBB) and 2-ethylhexyl 3,4,5,6-tetrabromophthalate (TBPH), were found above detection limits at different monitoring sites in MA and ME (Table 12).
- TBB was detected more frequently than TBPH, which was found in one mussel tissue sample from the Kennebec Perkins Island (MEKN) site in ME (Figure 12).
- With a calculated 15.8% frequency of detection Gulf-wide (Table 12), TBB was found at six monitoring sites; three (BHDI, BHHD, DBCI) in MA and three (MEKN, MEPR, MADI) in ME.
- The magnitude of the AFR contaminants detected varied greatly. A maximum concentration of 3.27 ng/g ww for TBB was recorded in the mussel sample from the MADI site in MA. TBPH was found at a concentration of 0.73 ng/g ww in mussels from the MEKN site in ME (Appendix 2).
- Our land-use assessment indicated that AFR contaminants were primarily located in coastal zone with land-use categorized as open-water or undeveloped (Table 4). Only site, MEPR, was associated with the developed land-use category. However, four of the six sites with detected AFR were located near urban areas such as the Boston and Portland harbors.

Site BBCC. Credit: NOAA

Jurisdiction-specific assessments: MASSACHUSETTS SUMMARY

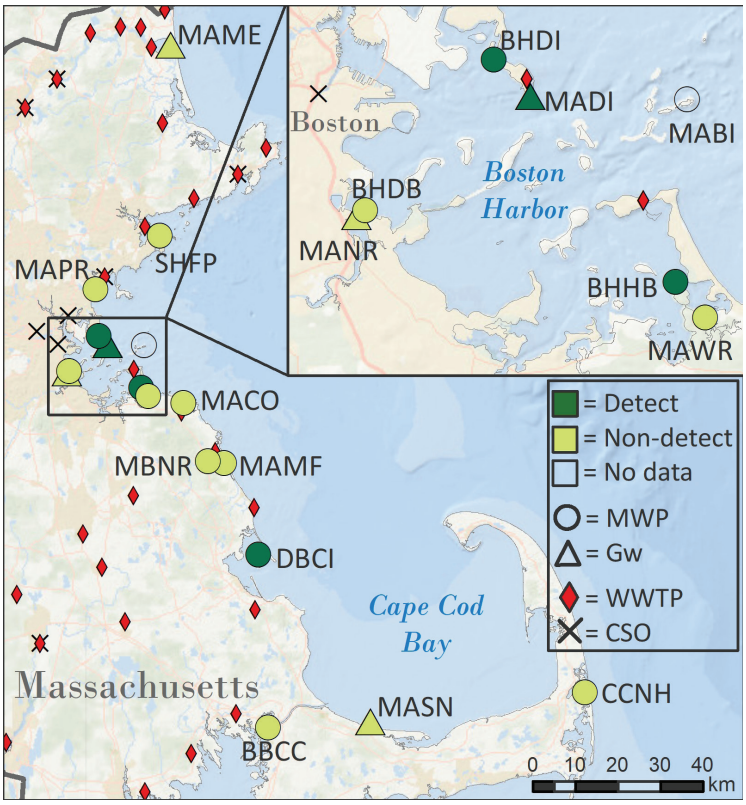


Figure 13. Map of MA jurisdiction highlighting location of sites with AFR detection in mussel tissue.

Table 14. AFR compounds frequency of detection in mussel tissue from MA jurisdiction (n = 16).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
TBB	4	16	25.0

Summary of AFRs in Massachusetts

- A total of 16 mussel tissue samples were tested for AFR contaminants in MA (Figure 13).
- Of the nine AFR contaminants tested, 2-ethylhexyl tetrabromobenzoate (TBB) was the only AFR detected in MA (Table 14).
- TBB was detected at four sites in MA, including the Druxbury Bay Clarks Island (DBCI), Boston Harbor Hingham Bay (BHHB), Boston Harbor Deer Island (BHDI), and Deer Island (MADI) (Figure 13).
- The maximum TBB concentration of 3.27 ng/g ww was detected at the Deer Island (MADI) site (Figure 12, Appendix 2).
- Concentrations of 1.56, 1.27 and 0.59 ng/g ww were recorded at the Druxbury Bay Clarks Island (DBCI), Boston Harbor Hingham Bay (BHHB), Boston Harbor Deer Island (BHDI) sites respectively (Appendix 2).
- The four monitoring sites with detected TBB contaminant were either located in Boston Harbor (BHHB, BHDI, and MADI) or within a semi-enclosed bay (DBCI), which might have influenced the accumulation of the AFR contaminants in mussel tissue.

Jurisdiction-specific assessments: NEW HAMPSHIRE SUMMARY

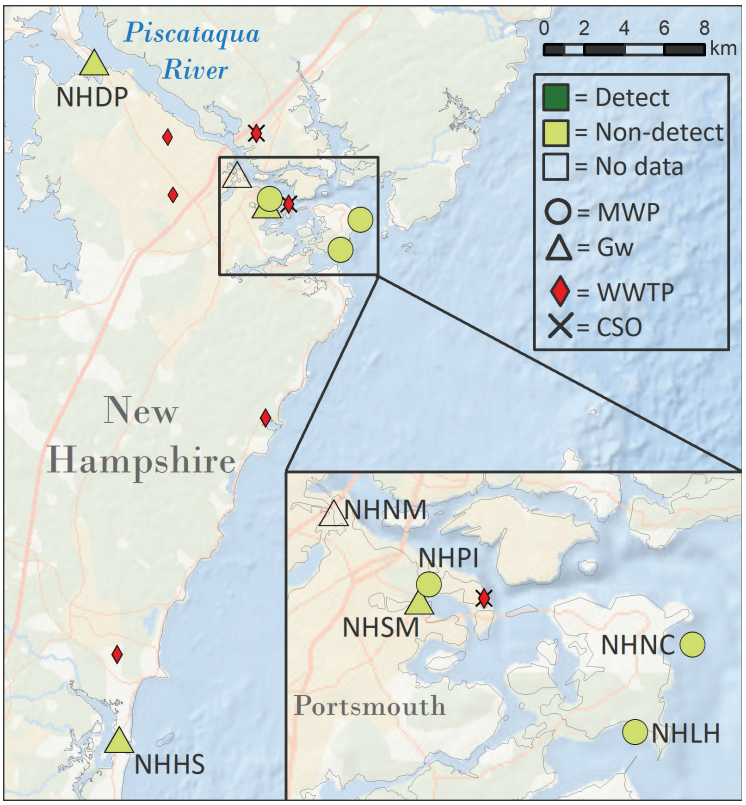


Figure 14. Map of NH jurisdiction highlighting location of sites with AFR detection in mussel tissue.

Summary of AFRs in New Hampshire

- Mussel tissue samples from six monitoring sites in NH were measured for AFR contaminants.
- None of the four AFR contaminants tested was detected in the NH jurisdiction (Figure 14).


Site MBNR. Credit: NOAA

Jurisdiction-specific assessments: MAINE SUMMARY

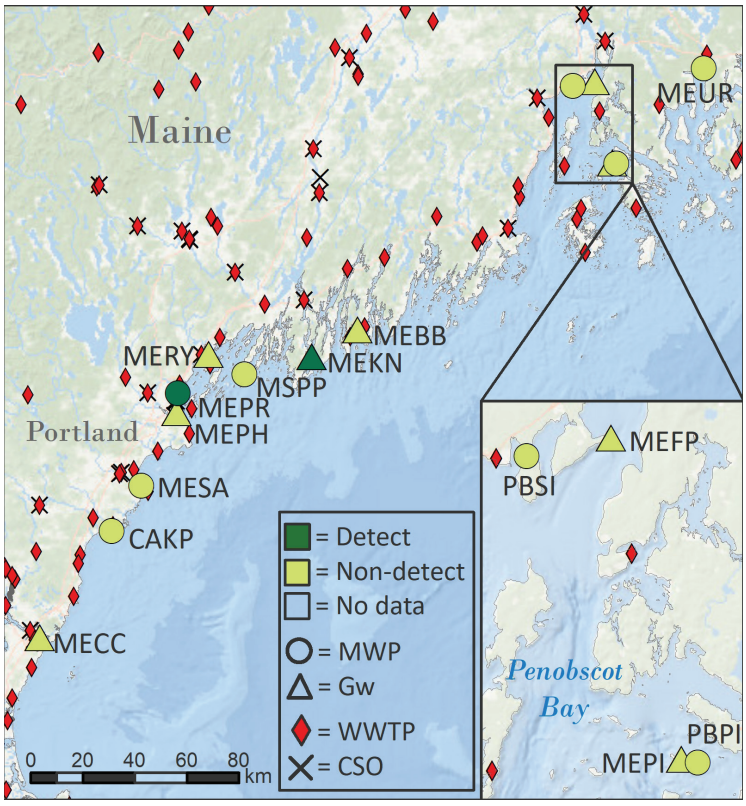


Figure 15. Map of ME jurisdiction highlighting location of sites with AFR detection in mussel tissue.

Table 15. AFR compounds frequency of detection in mussel tissue from ME jurisdiction (n = 14).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
TBB	2	14	14.3
TBPH	1	14	7.1

Summary of AFRs in Maine

- A total of 14 mussel tissue samples were tested for AFR contaminants in ME (Figure 15).
- Two of the nine AFR contaminants tested, 2-ethylhexyl tetrabromobenzoate (TBB) and 2-ethylhexyl 3,4,5,6-tetrabromophthalate (TBPH), were detected in mussel tissue from ME (Table 15).
- The TBB contaminant was detected at the Kennebec Perkins Island (MEKN) and the Presumpscot River (MEPR) sites at concentrations of 1.91 and 0.55 ng/g ww respectively (Figure 12, Appendix 2).
- TBPH was found at a concentration of 0.73 ng/g ww in mussels from the Kennebec Perkins Island (MEKN) site (Figure 12, Appendix 2).
- The two sites in ME, MEKN and MEPR, with detected AFR contaminants were located in coastal watersheds classified as undeveloped or open-water and developed respectively (Table 4). However, the presence of AFR contaminants at detectable levels may have resulted from discharges from wastewater treatment plants and combined sewer outfalls in the area or higher up in the watershed (Figure 15).

Jurisdiction-specific assessments: Nova Scotia Summary

Figure 16. Map of NS jurisdiction highlighting location of sites with AFR detection in mussel tissue.

Summary of AFRs in Nova Scotia

- Mussel tissue samples from two monitoring sites in NS were measured for AFR contaminants.
- None of the AFR contaminants tested was detected in the NS jurisdictions (Figure 16).

Site PBPI. Credit: NOAA

Current-Use Pesticides (CUPs)

Chemical Description

Unregulated or unmonitored contemporary contaminants are common and include current-use pesticides (CUPs) that are classified as organophosphate, neonicotinoids, pyrethroids, n-methyl carbamates, and insect growth regulator hormones. CUPs are generally a group of semi-volatile chemicals that span multiple chemical classes and can be analyzed concurrently. In this report, CUP chemicals include pesticides and their associated degradation products. These pesticides are typically more water-soluble than the legacy organochlorine pesticides and often do not bioaccumulate in organisms. It has been estimated that in 2007, over 565 million kg of current-use pesticides were used in the USA (EPA, 2011). Among pesticides, herbicides accounted for 40% of total usage, and insecticides 17% (EPA, 2011). While agriculture application accounts for over 60% of pesticides used, urban usage is increasing (EPA, 2011). Pesticides enter the environment seasonally through surface run-off, pesticide drift, direct discharge and through atmospheric long-range transport (USGS, 1999; Federighi, 2008).

The list of CUP chemicals measured in this study is restricted by available analytical methods for the chemicals identified in Table 16. SGS AXYS Analytical Services LTD. conducted these measurements. The analytical methods are proprietary and confidential but generally detect a group of semi-volatile chemicals that span multiple chemical classes. Hence, only the method name (MLA-035 REV.07.04) is mentioned in this document, along with contact information (SGS AXYS Analytical Services LTD., 2045 Mills Road W., Sidney, BC, Canada, V8L 5X2. Tel. (250) 655-5800, fax (250) 655-5811) for further references.

Table 16. CUP compounds tested.

Chemical name	Application
Ametryn	herbicide used to control broadleaf and grass weeds in fields planted with field corn, pineapple, and sugarcane
Atrazine	herbicide, used to control pre- and postemergence broadleaf weeds in crops
Azinphos-Methyl	broad spectrum organophosphate acetylcholinesterase inhibitor insecticide
Captan	fungicide
Chlorothalonil	broad spectrum non-systemic fungicide
Chlorpyrifos	organophosphate insecticide, acaricide and miticide used primarily to control a number of pests on food and feed crops
Chlorpyrifos-Methyl	organophosphate insecticide, used against a wide range of insects and pests, especially in grain storage
Chlorpyrifos-Oxon	chlorpyrifos metabolite, acts as an acetylcholinesterase inhibitor

Chemical Description

Table 16 (cont). CUP compounds tested.

Chemical name	Application
Cyanazine	herbicide
Cypermethrin	insecticide, used in large-scale commercial agricultural applications
Dacthal	pre-emergent herbicide, used to kill grass and many common weeds
Desethylatrazine	herbicide, a breakdown product of atrazine
Diazinon	nonsystemic organophosphate insecticide, formerly used to control cockroaches, silverfish, ants, and fleas in residential areas
Diazinon-Oxon	nonsystemic organophosphate insecticide, formerly used to control cockroaches, silverfish, ants, and fleas in residential areas
Dimethoate	organophosphate acetylcholinesterase inhibitor, used as an insecticide and acaricide
Disulfoton	organophosphate acetylcholinesterase inhibitor, used as an insecticide
Disulfoton Sulfone	organophosphate acetylcholinesterase inhibitor, used as an insecticide
Ethion	organophosphate insecticide
Fenitrothion	phosphorothioate (organophosphate) insecticide
Fonofos	organothiophosphate insecticide, primarily used on corn
Hexazinone	organic compound, used as a broad spectrum herbicide
Malathion	pesticide, widely used in agriculture and residential landscaping
Methoxychlor	insecticide, used to protect crops, ornamentals, livestock, and pets
Metribuzin	herbicide, used both pre- and post-emergence in crops including soy bean, potatoes, tomatoes and sugar cane; acts by inhibiting photosynthesis
Parathion-Ethyl	organothiophosphate insecticide, known as “Folidol”
Parathion-Methyl	insecticide, used on crops (cotton)
Permethrin	medication and insecticide; medication used to treat scabies and lice; insecticide sprayed on clothing or mosquito nets
Perthane	insecticide
Phosmet	non-systemic organophosphate insecticide, used on plants and animals
Pirimiphos-Methyl	phosphorothioate, used as an insecticide
Quintozene	fungicide
Simazine	herbicide of the triazine class, used to control broad-leaved weeds and annual grasses
Tecnazene	fungicide

Summary of CUPs in mussel tissue

- CUP contaminants were measured in 40 mussel tissue samples (17 in MA, 7 in NH, 14 in ME and 2 in NS) (there was not enough sample to test NSFI).
- Among the 33 CUP contaminants tested (Table 14), none were found at above the detection limit.

Per- and Polyfluoroalkyl Substances (PFASs)

Chemical Description

Per- and polyfluoroalkyl substances (PFASs) are a group of fluorine-containing compounds used in industrial processes related to surface protection/coatings, fire fighting foam, insecticides and commercial polymer manufacturing. Typically, PFASs enter the aquatic environment through aqueous industrial effluent from fire training/fire response sites, industrial sites, wastewater treatment plants and runoff from the land application of contaminated biosolids (ATSDR, 2018). This class of chemicals appears to accumulate in the environment, and because of their widespread use, they are becoming ubiquitous in sediment and tissue samples in coastal habitats (Chen et al., 2012; CDC, 2018). When they are taken up by organisms, PFASs are suspected to be endocrine disruptors and can cause developmental problems in animals (Grun and Blumberg, 2009). PFOS is one of the most toxic of the PFAS contaminants with available toxicological data. It has been linked to liver damage, cancer and immune system suppression in humans. Thus, this class of CECs has garnered increasing interest in the past 10-15 years. While the manufacturing of PFOS and PFOA has been phased out in the US, the EPA and several states have started developing health based guidelines for PFOS and PFOA (not detected in this study) in drinking water (Corder et al., 2018).

There are thousands of PFAS pollutants but only a few are becoming more routinely monitored in the environment. The MWP program is measuring 12 PFASs (Table 17) which are considered toxic and for which methodologies are well developed. In this study, SGS AXYS Analytical Services LTD. conducted measurement of PFASs in sediment and tissue samples. The analytical methods are proprietary and confidential. Hence, only the method name (MLA-043 REV.08.06) is mentioned in this document, along with contact information (SGS AXYS Analytical Services LTD., 2045 Mills Road W., Sidney, BC, Canada, V8L 5X2. Tel. (250) 655-5800, fax (250) 655-5811) for further references.

Table 17. PFAS compounds tested.

Chemical code	Chemical name
PFBS	Perfluorobutane sulfonate
PFDA	Perfluorodecanoic acid
PFDODA	Perfluorododecanoic acid
PFDS	Perfluorohexane sulfonate
PFHPA	Perfluoroheptanoic acid
PFHXA	Perfluorohexanoic acid
PFHXS	Perfluorohexane sulfonate
PFNA	Perfluorononanoic acid
PFOA	Perfluorooctanoate
PFOS	Perfluorooctane sulfonate
PFOSA	Perfluorooctane sulfonamide
PFUNDA	Perfluoroundecanoic acid

Presence and distribution of PFASs in mussel tissue: GULF-WIDE ASSESSMENT

Table 18. PFAS Gulf-wide frequency of detection in mussel tissue.

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PFOSA	16	40	40
PFOA	1	40	2.5
PFOS	1	40	2.5
Compound Class Total	18	480	3.8

Number of compounds detected:
3/12

Number of sites with detects:
16/40

Most detected compound:
PFOSA

Table 19. PFAS number of detects in mussel tissue at each site.

Site	State	Number of Detects	Number of Compounds Analyzed
MANR	MA	2	12
MEBB	ME	2	12
CCNH	MA	1	12
BBCC	MA	1	12
MACO	MA	1	12
MAWR	MA	1	12
MABI	MA	1	12
MADI	MA	1	12
MAME	MA	1	12
MECC	ME	1	12
MEPH	ME	1	12
MEKN	ME	1	12
NHNC	NH	1	12
NHSM	NH	1	12
NHLH	NH	1	12
NHNM	NH	1	12

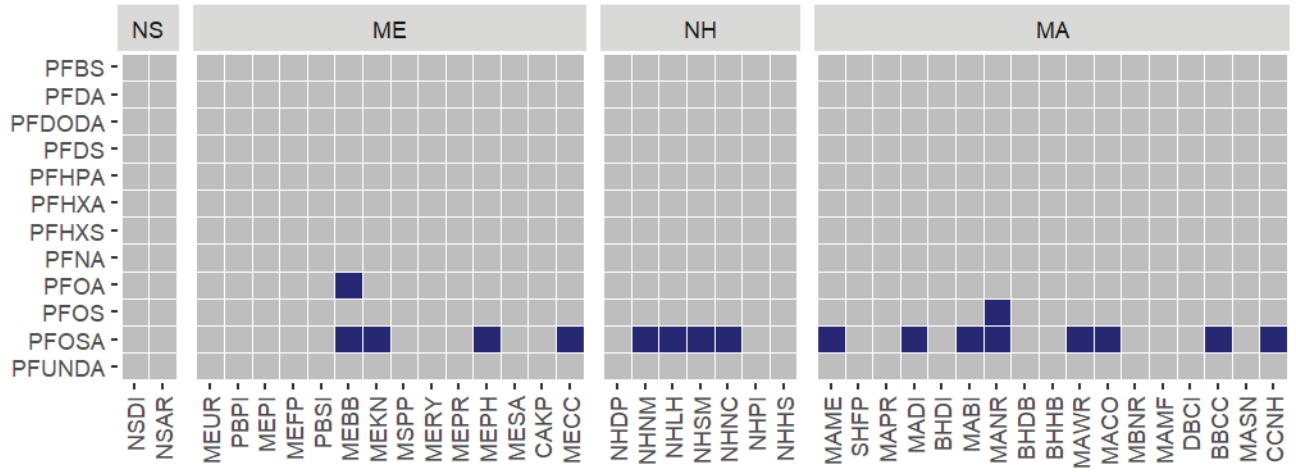


Figure 17. Distribution map showing presence (■) and absence (■) of PFASs measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

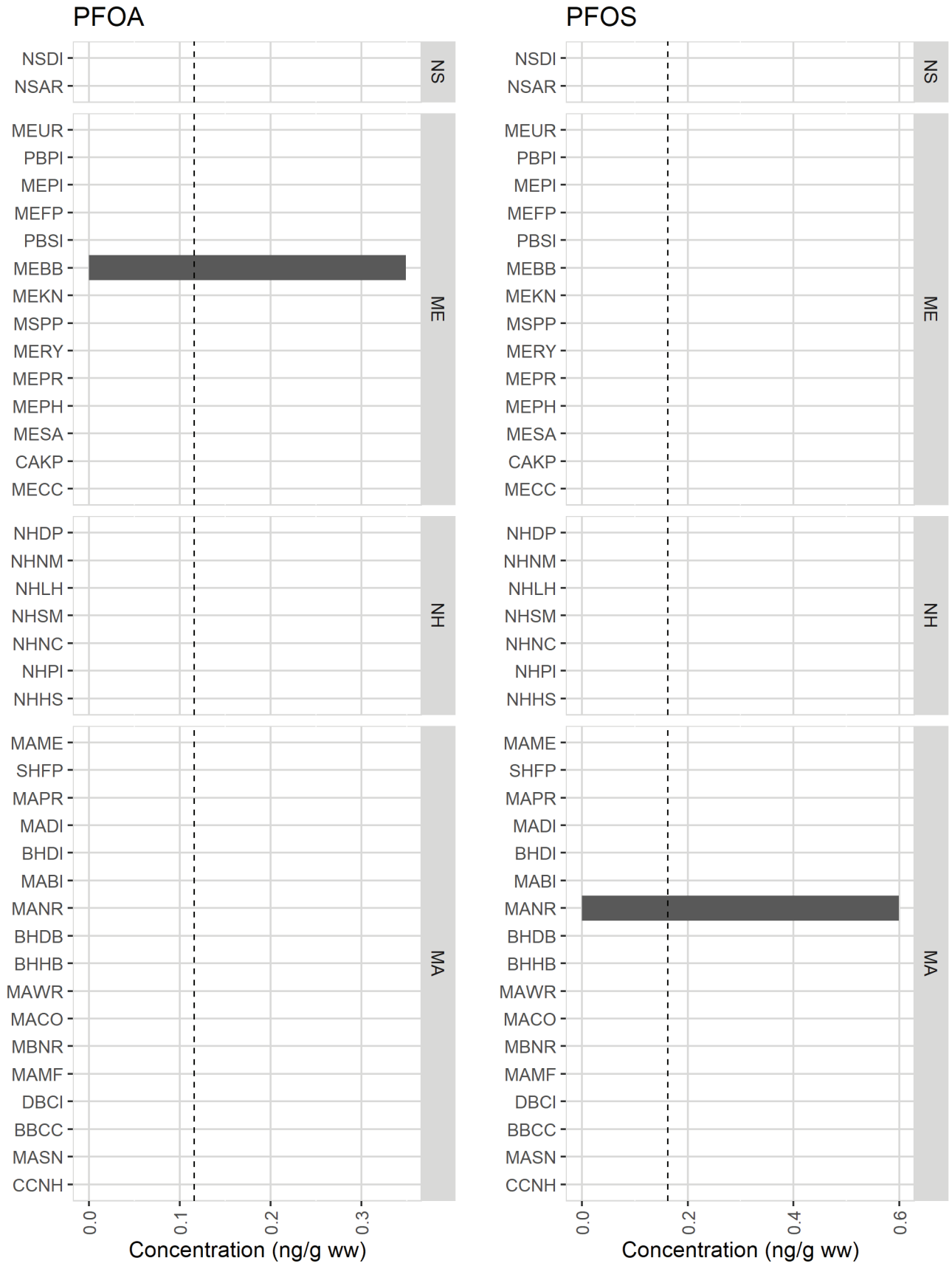
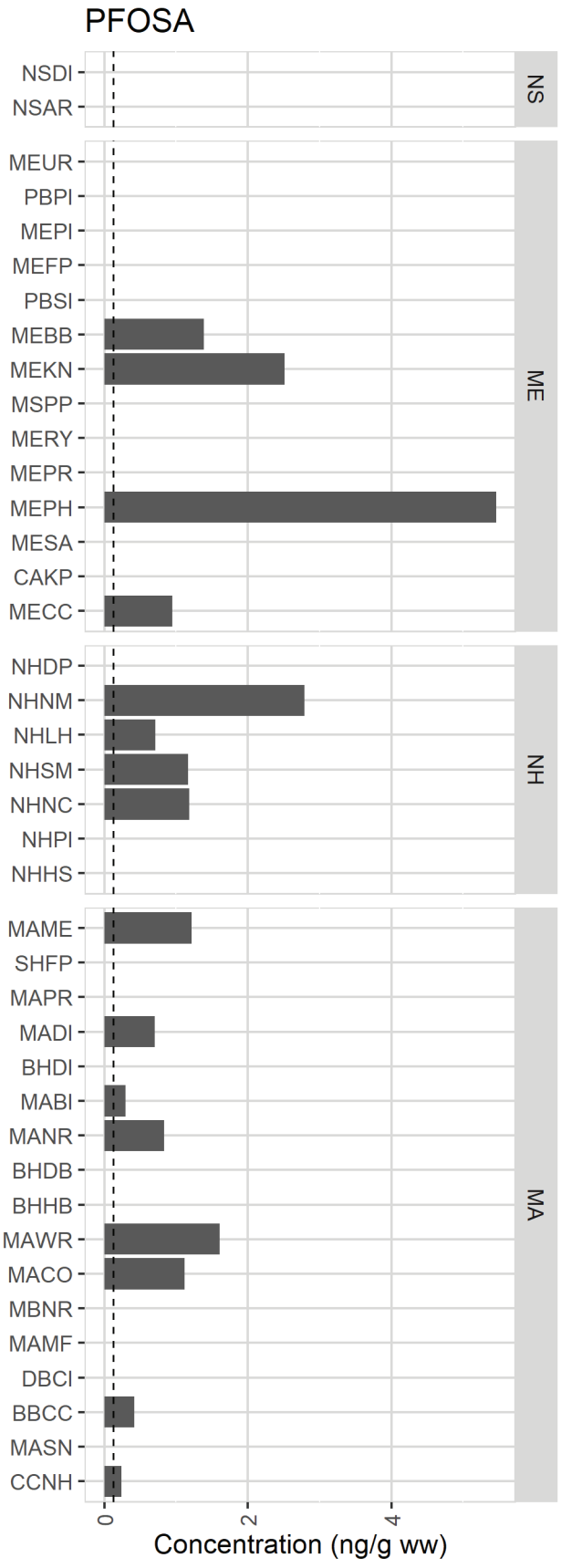


Figure 18. Bar graphs showing magnitude of PFASs detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

Gulf-wide Assessment



Summary of PFASs in mussel tissue

- Due to insufficient sample mass at NSFI, PFAS contaminants were measured in 40 mussel tissue samples (17 in MA, 14 in ME, 7 in NH and 2 in NS).
- Three of the 12 targeted PFAS contaminants, perfluorooctanoate (PFOA), perfluorooctane sulfonate (PFOS), and perfluorooctane sulfonamide (PFOSA) were found at above detection limits at different locations across the Gulf of Maine (Figure 17).
- Perfluorooctane sulfonamide (PFOSA) was the most frequently detected of the PFAS contaminants with estimated 40% frequency of detection Gulf-wide (Table 18). Perfluorooctane sulfonamide (PFOSA) was found at 16 monitoring sites across MA, NH and ME (Figure 17).
- Perfluorooctane sulfonate (PFOS) and perfluorooctanoate (PFOA) were each found at a single site in MA and ME respectively (Figure 17).
- The magnitude of PFAS contaminants detected varied greatly in mussel tissue across the Gulf of Maine. A maximum concentration of 5.46 ng/g ww was recorded for PFOSA at the MEPH in Maine (Figure 18, Appendix 4). Perfluorooctane sulfonate (PFOS) found at the MANR site in MA had a concentration of 0.60 ng/g ww, while perfluorooctanoate (PFOA) found at the MEBB site in ME had a concentration of 0.35 ng/g ww.
- No PFAS contaminants were detected at the two Nova Scotia sites.
- Based on our land-use assessment, only half of the sites with detected PFAS compounds between ME (MEPH, MECC), MA (MACO, MANR, BBCC, MAWR) and NH (NHNM, NHSM) were located in the developed land-use category (Table 4). The remaining sites in NH (NHNC, NHLH), ME (MEBB, MEKN) and MA (CCNH, MABI, MADI, MAME) with detectable levels of PFASs were associated with open-water or undeveloped land-use categories (Table 4).
- PFAS detection frequencies and PFOSA concentrations were positively correlated with percent impervious surface in a 1 km buffer (p=.015, rho=0.39; p=.022, rho=0.37). PFOSA concentrations were higher at sites in the developed land-use category than open-water sites in the 2 and 3 km buffers (p=.032 and p=.049, respectively). (Appendix 6)

Figure 18. Bar graphs showing magnitude of PFAS contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

Jurisdiction-specific assessments: MASSACHUSETTS SUMMARY

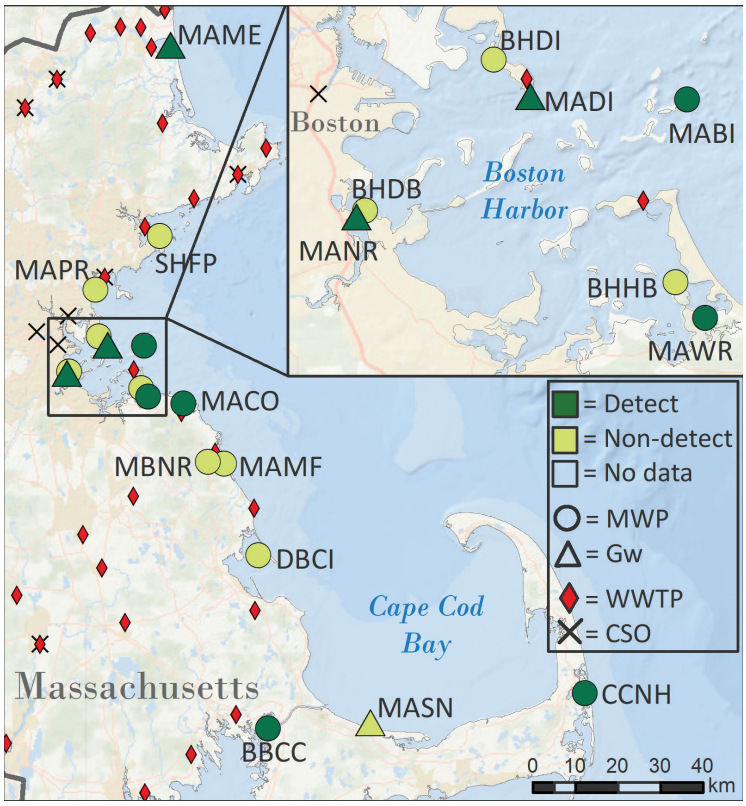


Figure 19. Map of MA jurisdiction highlighting location of sites with PFAS detection in mussel tissue.

Table 20. PFAS compounds frequency of detection in mussel tissue from MA jurisdiction (n = 17).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PFOSA	8	17	47.1
PFOS	1	17	5.9

Summary of PFASs in Massachusetts

- A total of 17 mussel tissue samples were tested for PFASs in MA (Figure 19).
- Two out of 12 PFAS contaminants tested were found in MA, including perfluorooctane sulfonamide (PFOSA) and perfluorooctane sulfonate (PFOS) (Table 20).
- Perfluorooctane sulfonamide (PFOSA) was found at 8 of the 17 monitoring sites with a frequency of detection of 47.1% in MA (Table 20)
- In MA, PFOSA was found at the Buzzard Bay Cape Cod (BBCC), Nauset Harbor Cape Cod (CCNH), Boston Harbor Brewster Island (MABI), Cohassett (MACO), Deer Island (MADI), Merrimack River (MAME), Neponset River (MANR), and Weir River Estuary (MAWR) sites (Figure 19).
- The magnitude of PFOSA detected in MA varied from 0.24 ng/g ww at the CCNH site to a maximum concentration of 1.61 ng/g ww found in mussels from the MAWR site (Figure 15, Appendix 3).
- Perfluorooctane sulfonate (PFOS) was found at the concentration of 0.60 ng/g ww at the Neponset River (MANR) site only (Figure 19).
- The total concentration of detected PFAS contaminants in MA was 7.03 ng/g ww.
- In this study, MA monitoring sites with PFAS detects were associated with developed, undeveloped and open-water land-use categories (Table 4), however, results indicated that the majority of the mussel samples with detected PFAS contaminants were located in the Boston Harbor area.

Jurisdiction-specific assessments: NEW HAMPSHIRE SUMMARY

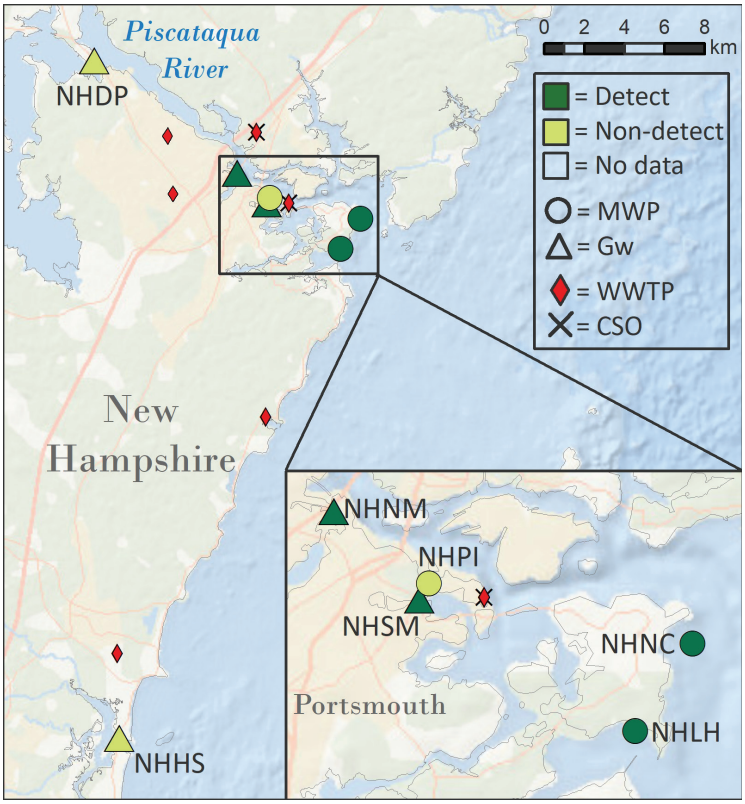


Figure 20. Map of NH jurisdiction highlighting location of sites with PFAS detection in mussel tissue.

Table 21. PFAS compounds frequency of detection in mussel tissue from NH jurisdiction (n = 7).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PFOSA	4	7	57.1

Summary of PFASs in New Hampshire

- Mussel tissue samples from seven monitoring sites in NH were measured for PFAS contaminants (Figure 20).
- Out of 12 PFAS contaminants tested, only perfluorooctane sulfonamide (PFOSA) was found in NH (Table 21).
- With a frequency of detection of 57.1%, PFOSA was found at four of the seven monitoring sites in NH including the Piscataqua River Little Harbor (NHLH), New Castle (NHNC), North Mill Pond (NHNM) and South Mill Pond (NHSM) sites (Figure 20).
- The magnitude of PFOSA detected in NH varied from 0.71 ng/g ww at the NHLH site to 2.79 ng/g ww at the NHNM site (Figure 18, Appendix 3).
- Based on our land-use assessment, two of the four sites with detects (NHNM, NHSM) were associated with developed land-use (Table 4). Furthermore, the sites with detected PFAS contaminants were along or in the delta area at the mouth of the Piscataqua River, which drains a watershed that harbors some wastewater treatment plants and combined sewer outfalls (Figure 20).

Jurisdiction-specific assessments: MAINE SUMMARY

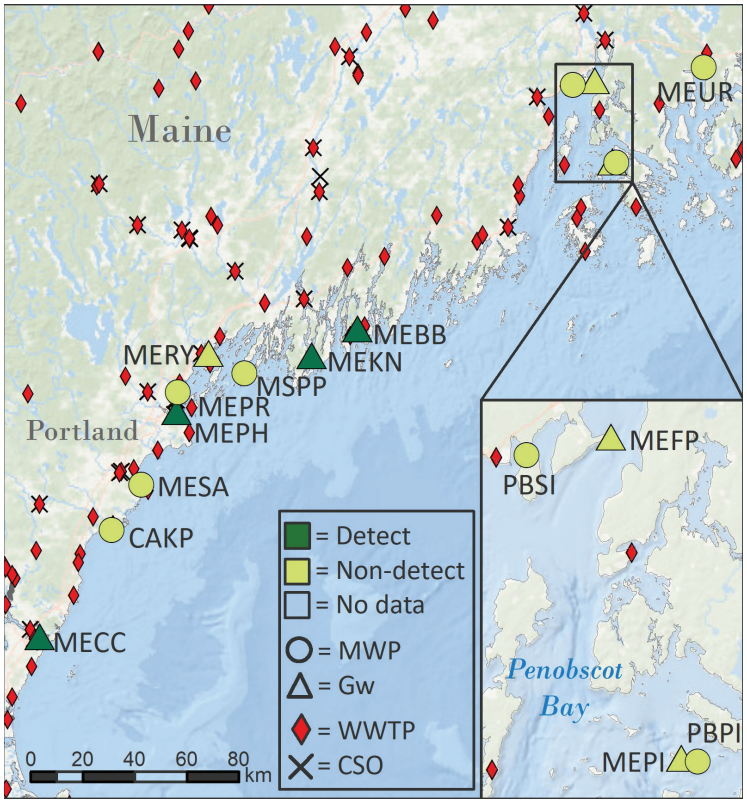


Figure 21. Map of ME jurisdiction highlighting location of sites with PFAS detection in mussel tissue.

Table 22. PFAS compounds frequency of detection in mussel tissue from ME jurisdiction (n = 14).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PFOSA	4	14	28.6
PFOA	1	14	7.1

Summary of PFASs in Maine

- A total of 14 mussel tissue samples were tested for PFAS contaminants in ME (Figure 21).
- Two of the 12 PFAS contaminants tested were found in ME, perfluorooctane sulfonamide (PFOSA) and perfluorooctanoate (PFOA) (Table 22).
- With a frequency of detection of 28.6% PFOSA was found at four of the 14 monitoring sites in ME including the Boothbay Harbor (MEBB), Salmon Falls Clark Cove (MECC), Kennebec Perkins Island (MEKN) and Stroudwater-Force Portland Harbor (MEPH) monitoring sites (Table 22, Figure 21).
- The magnitude of PFOSA detected in ME varied from 1.39 ng/g ww at the MEBB site to 5.46 ng/g ww at the MEPH site (Figure 18, Appendix 3).
- Perfluorooctanoate (PFOA) was found with a concentration of 0.35 ng/g ww at the MEBB site only in ME (Figure 18, Appendix 3).
- Based on our land-use classification, PFAS contaminants were detected in both primarily developed (MEPH, MECC) and primarily undeveloped or open-water (MEKN, MEBB) land-use categories (Table 4). However, there is the potential for influences from water treatment plants and/or combined sewer outfalls at all of these sites whose discharges are likely to influence water quality in the area (Figure 21).

Jurisdiction-specific assessments: NOVA SCOTIA SUMMARY

Figure 22. Map of NS jurisdiction highlighting location of sites with PFAS detection in mussel tissue.

Summary of PFASs in Nova Scotia

- Mussel tissue samples from two monitoring sites in NS were measured for PFAS contaminants.
- None of the 12 PFAS contaminants tested were detected in the NS jurisdictions (Figure 22).

Pharmaceuticals and Personal Care Products (PPCPs)

Chemical Description

Environmental pharmaceuticals and personal care products (PPCPs) include a wide spectrum of therapeutic and consumer-use compounds such as prescription and over-the-counter medications, hormones, synthetic fragrances, detergents, disinfectants, insect repellants, and antimicrobial agents. In 2009, an estimated 3.9 billion prescriptions were written for the top 300 pharmaceuticals in the US (Lundy, 2010). Pharmaceutical companies produce over 50 million pounds of antibiotics annually in the United States with approximately 60% for human use and 40% for animal agriculture (Levy, 1998). There are numerous pathways by which PPCPs are introduced into the environment, although the primary routes include wastewater discharge after excretion or improper disposal of unused drugs (Daughton and Ternes, 1999). Because pharmaceuticals are designed with the intention of having a biological effect, the major concerns associated with PPCPs in the environment are their potential ecotoxicity and unintentional human health impacts. Potential impacts of PPCPs in the environment include abnormal physiological effects, impaired reproduction, and increased cancer rates (Boyd and Furlong, 2002). According to the US EPA, many CECs including PPCPs are suspected to be endocrine disruptors, which alter the normal functions of hormones resulting in a variety of health effects (Ankley et al., 2008). For this study a total of 121 individual PPCP compounds were tested in mussel tissues (Table 23a-g).

Pharmaceutical and personal care products represent a diverse class of emerging contaminants among which selected compounds are measured by the MWP. The PPCPs analyzed in this study are grouped by analytical methods identified as HormoneNEG, HormonePOS, PPCP-I, PPCP-III, PPCP-IV, PPCP-V and PPCP-VI (Table 23a-g). The analyses were conducted by the NCCOS' chemistry laboratory in Charleston, SC. Sample extraction, clean-up and quantitation procedures were based on modified EPA method 1694 (EPA, 2007) and methods described in Klosterhaus et al. (2013) and Apeti et al. (2018).

Chemical Description

Table 23a. HormoneNEG compounds tested (n=9).

Chemical code	General Use
17α-Dihydroequilin	Steroidal Estrogen
17α-estradiol	Steroidal Estrogen
17α-Ethynyl estradiol	Steroidal Estrogen
17β-estradiol	Steroidal Estrogen
Diethylstilbestrol	Nonsteroidal Estrogen
Equilenin	Steroidal Estrogen
Equilin	Steroidal Estrogen
Estriol	Steroidal Estrogen
Estrone	Steroidal Estrogen

Table 23b. HormonePOS compounds tested (n=8).

Chemical code	General Use
Allyl Trenbolone	Progestin
Androstenedione	Steroidal Androgen
Androsterone	Steroidal Androgen
Desogestrel	Progestin
Mestranol	Estrogen
Norgestrel	Progestin
Progesterone	Progestin
Testosterone	Steroidal Androgen

Table 23c. PPCP-I compounds tested (n=43).

Chemical code	General Use
Acetaminophen	Pain Reliever
Azithromycin	Antibiotic
Caffeine	Stimulant
Carbadox	Antibiotic
Carbamazepine	Anticonvulsant
Ciprofloxacin	Antibiotic
Clarithromycin	Antibiotic
Clinafloxacin	Antibiotic
Cloxacillin	Antibiotic
Dehydronifedipine	Cardiovascular

Table 23c (cont). PPCP-I compounds tested (n=43).

Chemical code	General Use
Digoxigenin	Steroid
Digoxin	Cardiovascular
Diltiazem	Cardiovascular
Diphenhydramine	Antihistamine
Enrofloxacin	Antibiotic
Erythromycin	Antibiotic
Flumequine	Antibiotic
Fluoxetine	Psychiatric
Lomefloxacin	Antibiotic
Miconazole	Antifungal
Norfloxacin	Antibiotic
Norgestimate	Progestin
Ofloxacin	Antibiotic
Ormetoprim	Antiprotozoal
Oxacillin	Antibiotic
Oxolinic acid	Antibiotic
Paraxanthine	Stimulant
Penicillin G	Antibiotic
Penicillin V	Antibiotic
Roxithromycin	Antibiotic
Sarafloxacin	Antibiotic
Sulfachloropyridazine	Antibiotic
Sulfadiazine	Antibiotic
Sulfadimethoxine	Antibiotic
Sulfamerazine	Antibiotic
Sulfamethazine	Antibiotic
Sulfamethizole	Antibiotic
Sulfamethoxazole	Antibiotic
Sulfanilamide	Antibiotic
Sulfathiazole	Antibiotic
Thiabendazole	Antifungal
Trimethoprim	Antibiotic
Tylosin	Antibiotic

Chemical Description

Table 23d. PPCP-III compounds tested (n=12).

Chemical code	General Use
2-Hydroxy-ibuprofen	Pain Reliever
Bisphenol-A	Plastic Additive
Furosemide	Fluid Reducer
Gemfibrozil	Cholesterol Reducer
Glipizide	Antidiabetic
Glyburide	Antidiabetic
Hydrochlorothiazide	High Blood Pressure
Ibuprofen	Pain Reliever
Naproxen	Pain Reliever
Triclocarban	Antibacterial
Triclosan	Antibacterial/ Antifungal
Warfarin	Cardiovascular

Table 23e. PPCP-IV compounds tested (n=14).

Chemical code	General Use
Albuterol	Cardiovascular
Amphetamine	Psychiatric
Atenolol	Cardiovascular
Atorvastatin	Cholesterol Reducer
Cimetidine	Acid Reducer
Clonidine	Cardiovascular
Codeine	Pain Reliever
Cotinine	Recreational Drug
Enalapril	Cardiovascular
Hydrocodone	Stimulant
Metformin	Antidiabetic
Oxycodone	Pain Reliever
Ranitidine	Acid Reducer
Triamterene	Diuretic

Table 23f. PPCP-V compounds tested (n=30).

Chemical code	General Use
10-hydroxy-amitriptyline	Psychiatric
Alprazolam	Psychiatric
Amitriptyline	Psychiatric
Amlodipine	Cardiovascular
Benzoylecgonine	Recreational Drug
Benztropine	Anti-Tremor
Betamethasone	Dermatitis
Cocaine	Recreational Drug
DEET	Insect Repellent
Diazepam	Psychiatric
Fluocinonide	Steroid
Fluticasone propionate	Steroid
Hydrocortisone	Steroid
Meprobamate	Psychiatric
Methylprednisolone	Steroid
Metprolol	Cardiovascular
N-Desmethyldiltiazem	Cardiovascular
Norfluoxetine	Psychiatric
Norverapamil	Cardiovascular
Paroxetine	Psychiatric
Prednisolone	Steroid
Prednisone	Steroid
Promethazine	Depressant
Propoxyphene	Pain Reliever
Propranolol	Cardiovascular
Sertraline	Psychiatric
Simvastatin	Cholesterol
Theophylline	Asthma
Valsartan	High Blood Pressure
Verapamil	Cardiovascular

Table 23g. PPCP-VI compounds tested (n=5).

Chemical code	General Use
Busulfan	Cardiovascular
Citalopram	Psychiatric
Clotrimazole	Cardiovascular
Etoposide	Cholesterol Reducer
Venlafaxine	Acid Reducer

Presence and distribution of PPCPs in mussel tissue: GULF-WIDE ASSESSMENT

Table 24. PPCP compounds Gulf-wide frequency of detection in mussel tissue.

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
DEET	35	40	87.5
Sertraline	17	40	42.5
Diphenhydramine	12	40	30
Ranitidine	9	40	22.5
Triamterene	5	40	12.5
Hydrocortisone	3	40	7.5
Testosterone	3	40	7.5
Triclocarban	3	40	7.5
17β-estradiol	2	40	5
Miconazole	2	40	5
Propranolol	2	40	5
Amitriptyline	1	40	2.5
Androstenedione	1	40	2.5
Atenolol	1	40	2.5
Azithromycin	1	40	2.5
Benzoylecgonine	1	40	2.5
Benztropine	1	40	2.5
Caffeine	1	40	2.5
Carbadox	1	40	2.5
Carbamazepine	1	40	2.5
Cimetidine	1	40	2.5
Citalopram	1	40	2.5
Cocaine	1	40	2.5
Cotinine	1	40	2.5
Diazepam	1	40	2.5
Fluoxetine	1	40	2.5
Meprobamate	1	40	2.5
Mestranol	1	40	2.5
Metprolol	1	40	2.5
Norgestrel	1	40	2.5
Propoxyphene	1	40	2.5
Compound Class Total	113	4838	2.3

Table 25. PPCP compounds number of detects in mussel tissue at each site.

Site	State	Number of Detects	Number of Compounds Analyzed
MANR	MA	6	121
BHDB	MA	5	120
BHDI	MA	5	121
NHHS	NH	5	121
NHSM	NH	5	121
NHDP	NH	5	121
MECC	ME	5	121
MAPR	MA	4	121
SHFP	MA	4	120
MAME	MA	4	121
NHPI	NH	4	121
NHNM	NH	4	121
MESA	ME	4	121
MEPR	ME	4	121
MEKN	ME	4	121
MADI	MA	3	121
NHNC	NH	3	121
MEPH	ME	3	121
MERY	ME	3	121
MSPP	ME	3	121
MEBB	ME	3	121
PBPI	ME	3	121
BBCC	MA	2	121
MBNR	MA	2	121
BHHB	MA	2	121
MABI	MA	2	121
PBSI	ME	2	121
MEFP	ME	2	121
MEUR	ME	2	121
CCNH	MA	1	121
MASN	MA	1	121
DBCI	MA	1	121
MAMF	MA	1	121
MACO	MA	1	121
MAWR	MA	1	121
NHLH	NH	1	121
CAKP	ME	1	121
MEPI	ME	1	121
NSDI	NS	1	121

Number of compounds detected:
31/121

Number of sites with detects:
39/40

Most detected compound:
DEET

GULF-WIDE ASSESSMENT

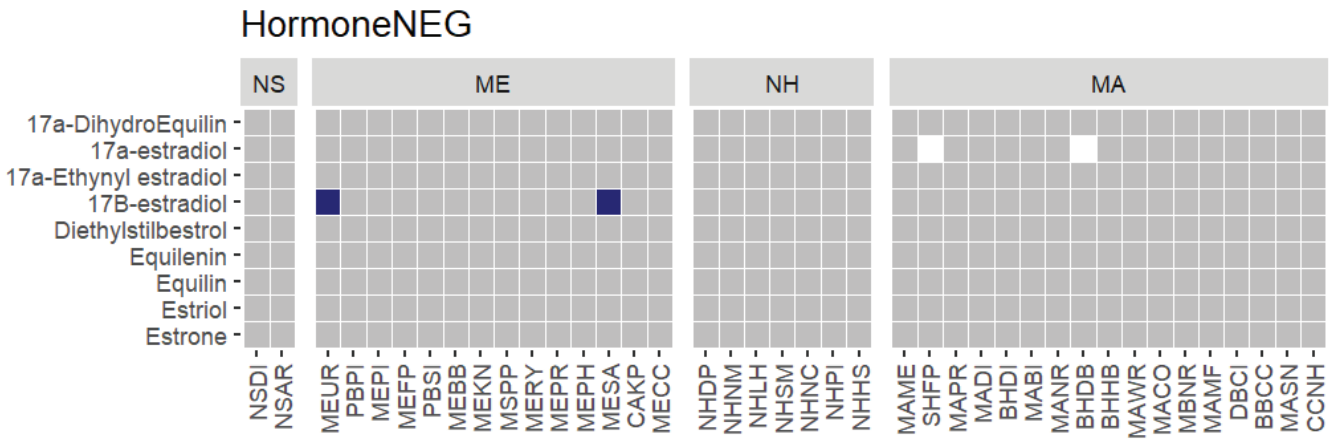


Figure 23a. Distribution map showing presence (■) and absence (■) of HormoneNEG compounds measured in mussel tissues from the Gulf of Maine. White squares represent analyte/site combinations for which results are not available. Sites are listed geographically from north to south, following the coastline.

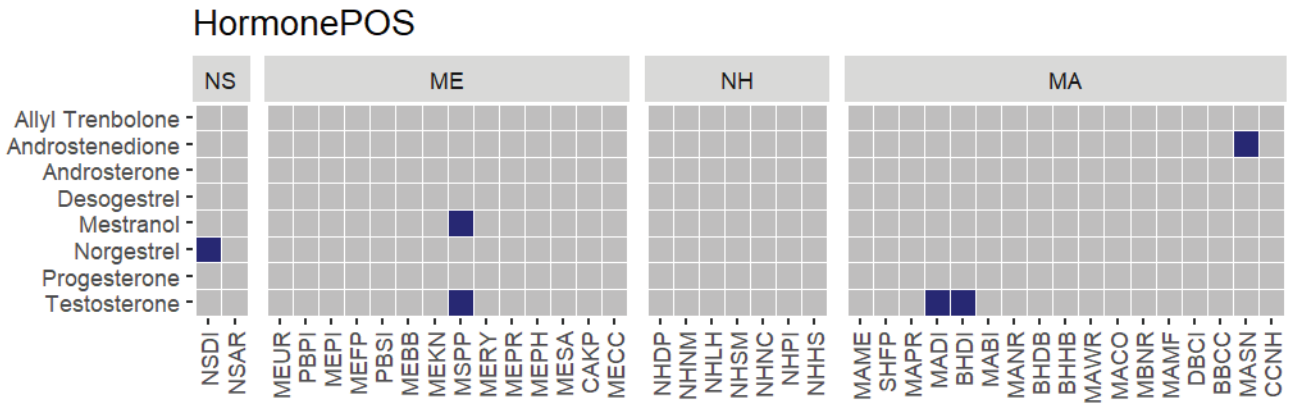


Figure 23b. Distribution map showing presence (■) and absence (■) of HormonePOS compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

Gulf-wide Assessment

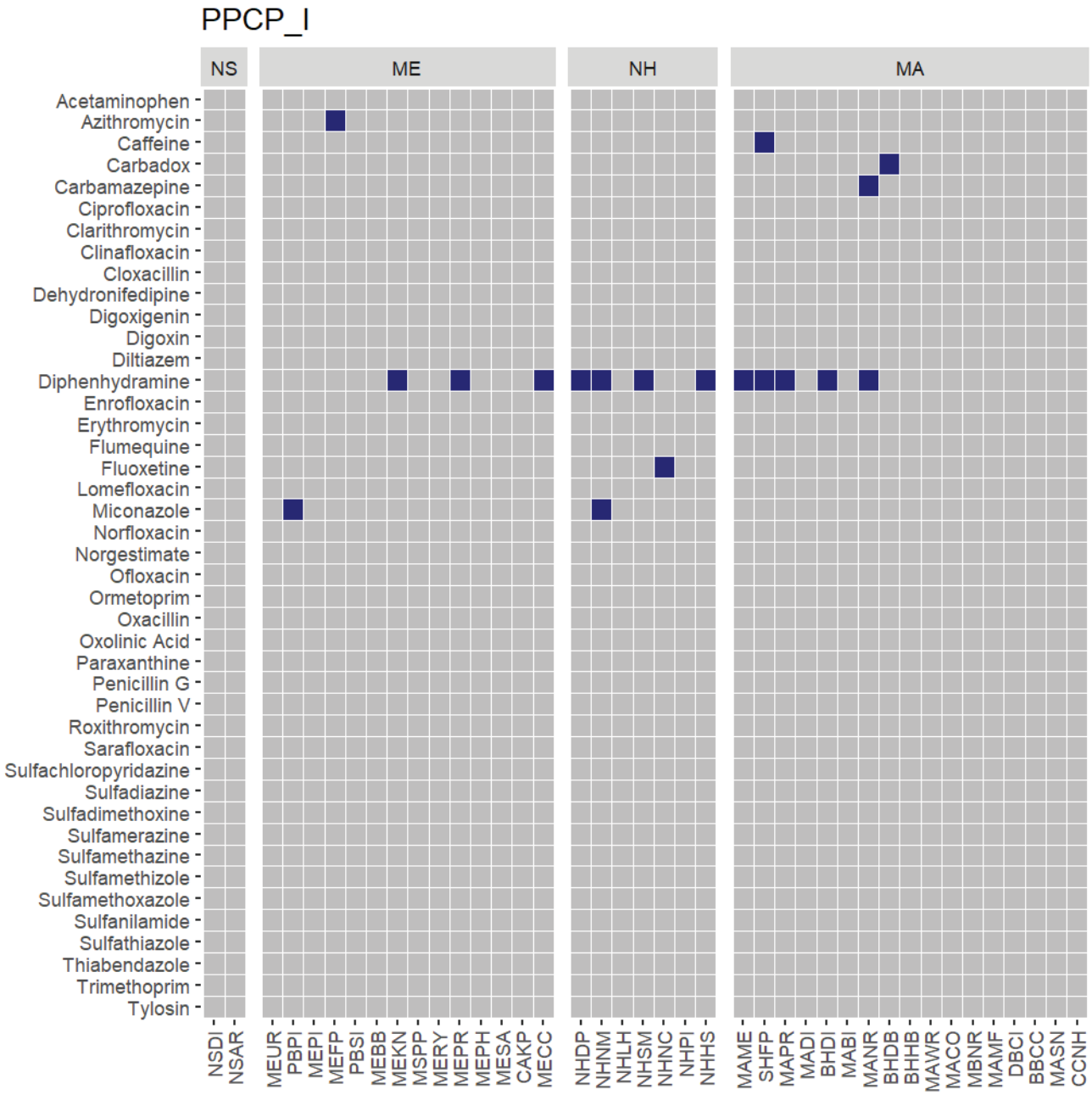


Figure 23c. Distribution map showing presence (■) and absence (■) of PPCP-I compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

Gulf-wide Assessment

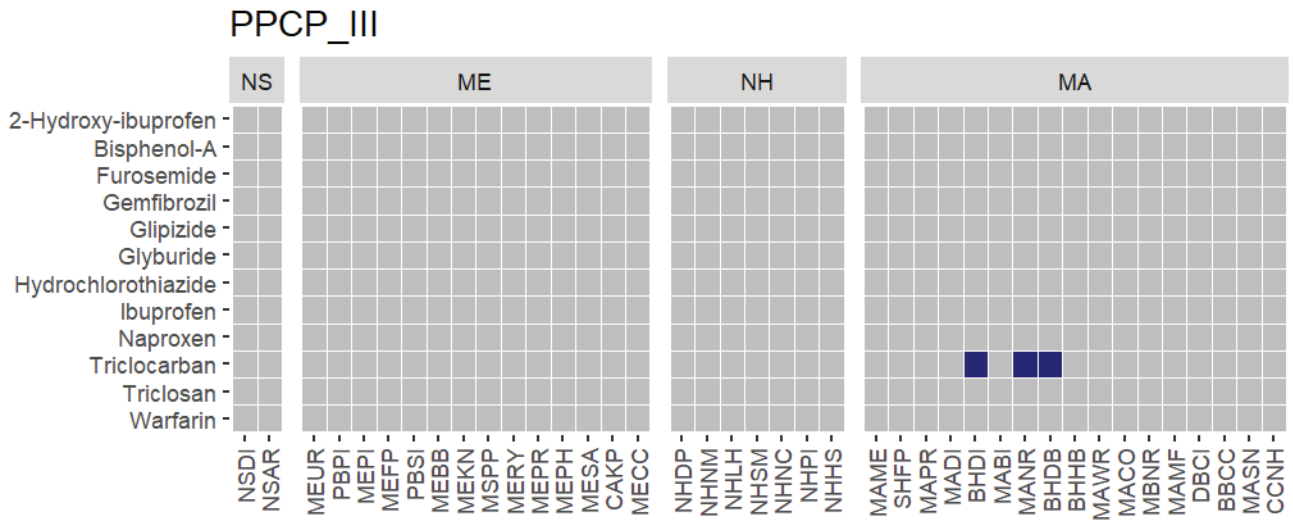


Figure 23d. Distribution map showing presence (■) and absence (■) of PPCP-III compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

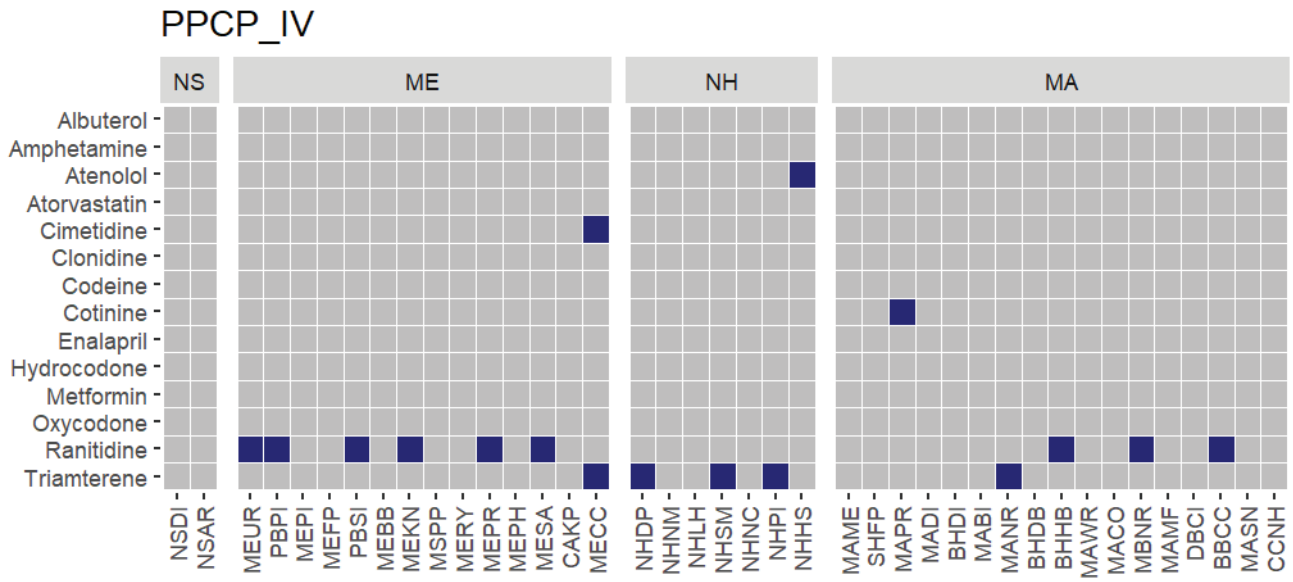


Figure 23e. Distribution map showing presence (■) and absence (■) of PPCP-IV compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

Gulf-wide Assessment

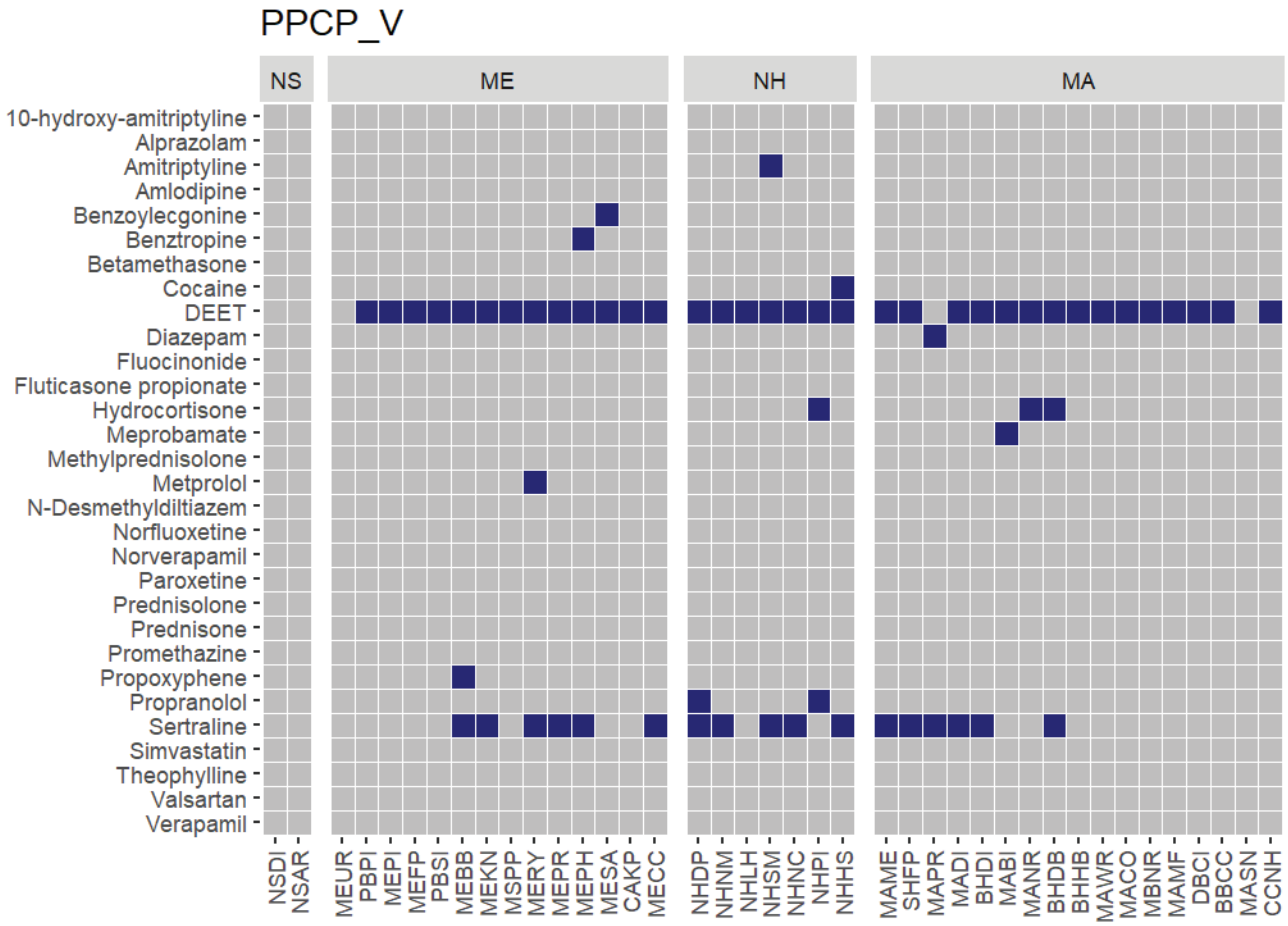


Figure 23f. Distribution map showing presence (■) and absence (■) of PPCP-V compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

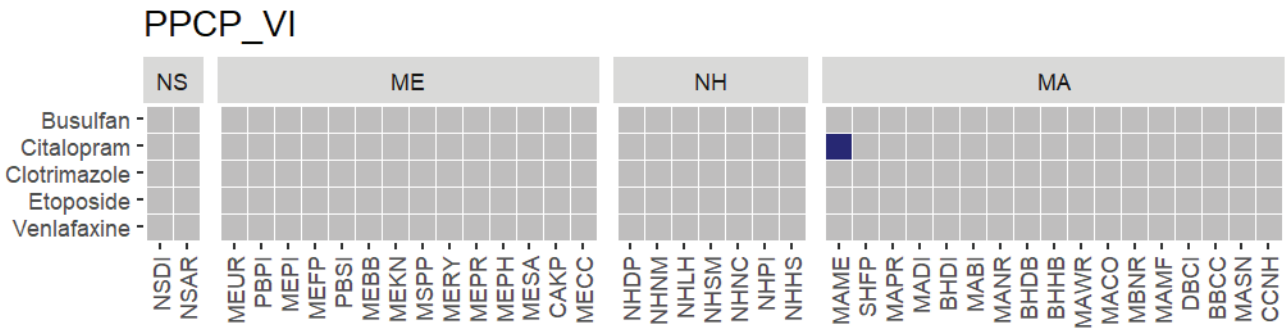


Figure 23g. Distribution map showing presence (■) and absence (■) of PPCP-VI compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

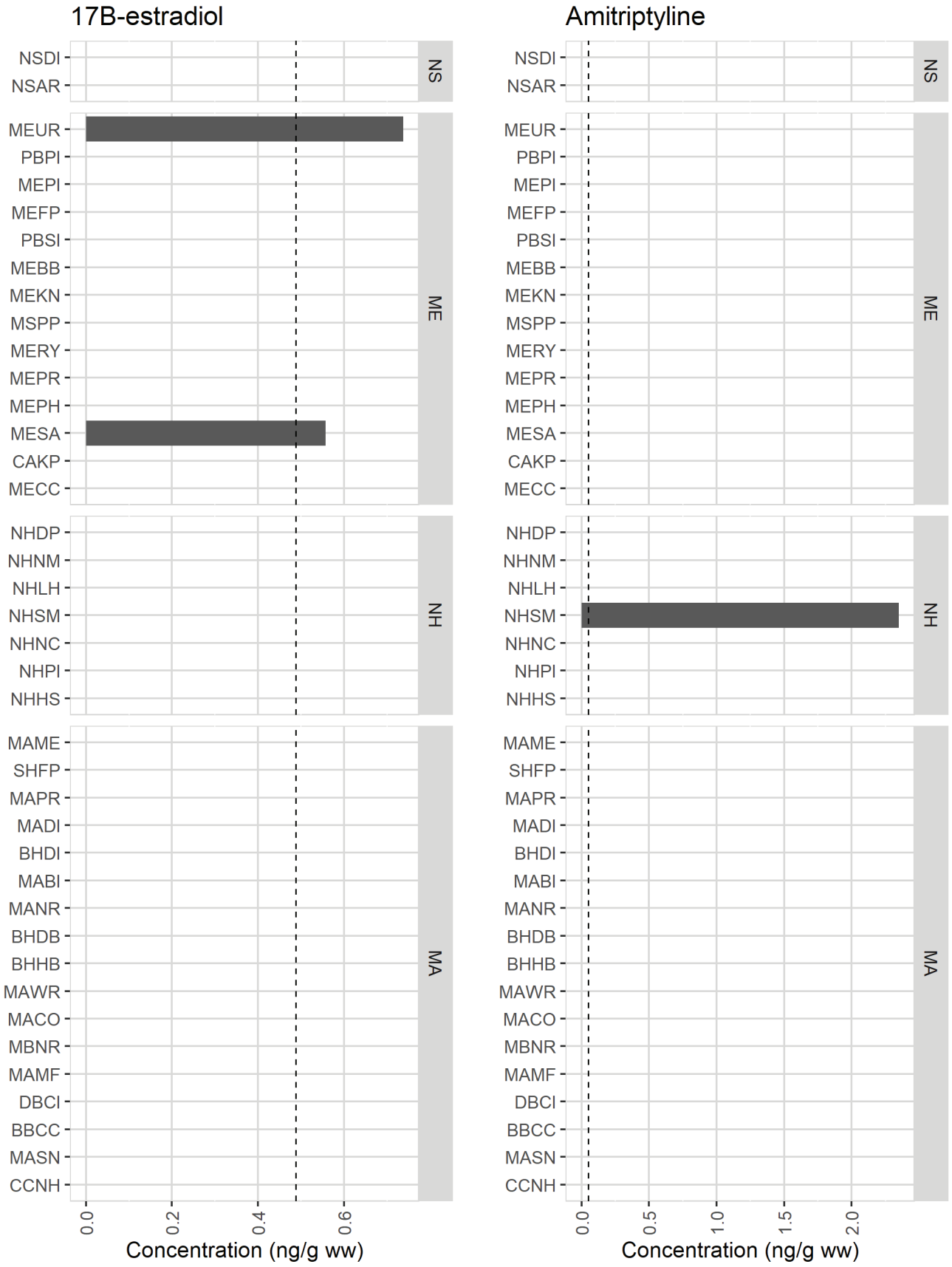
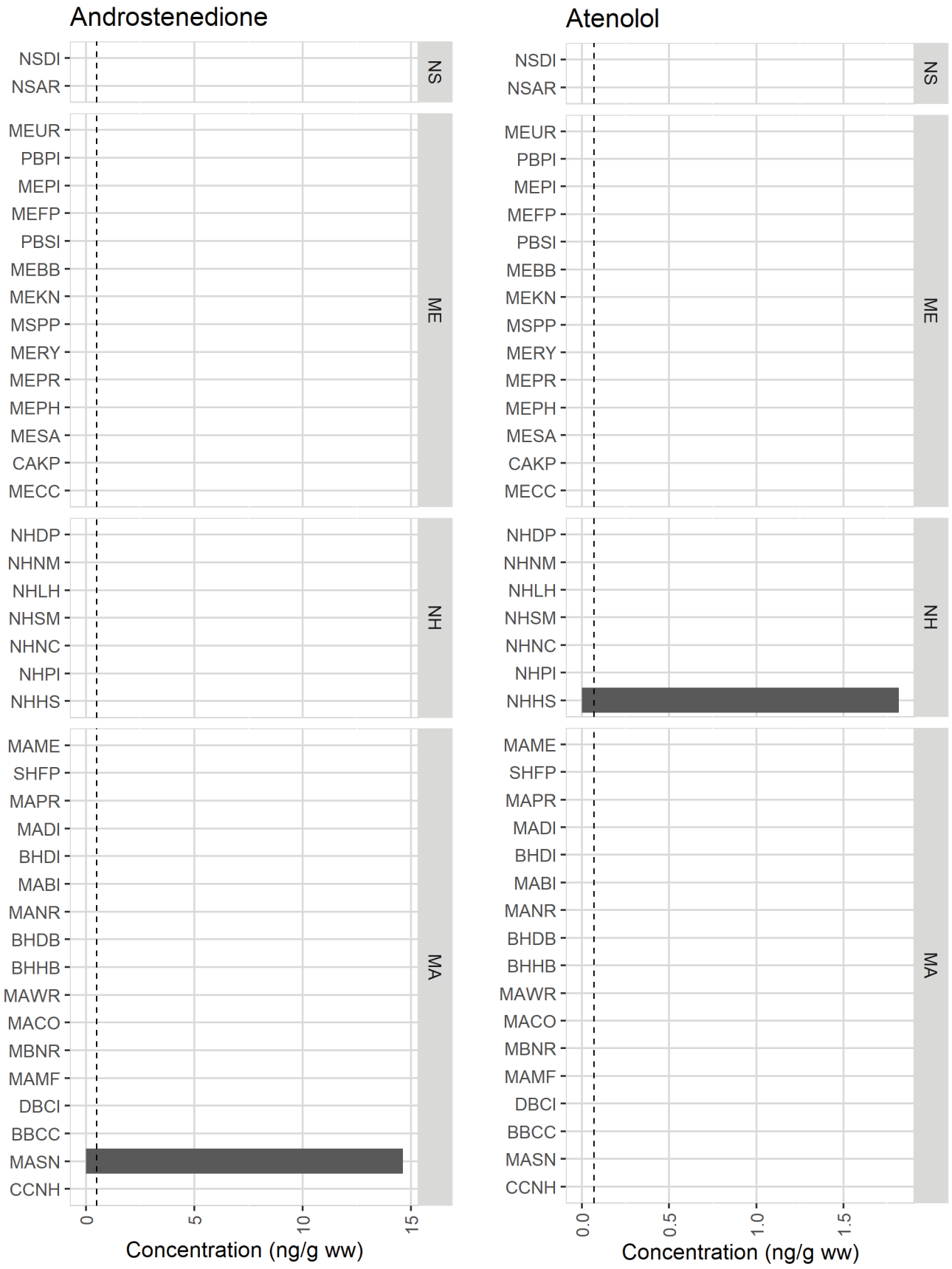


Figure 24. Bar graphs showing magnitude of PPCP contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT



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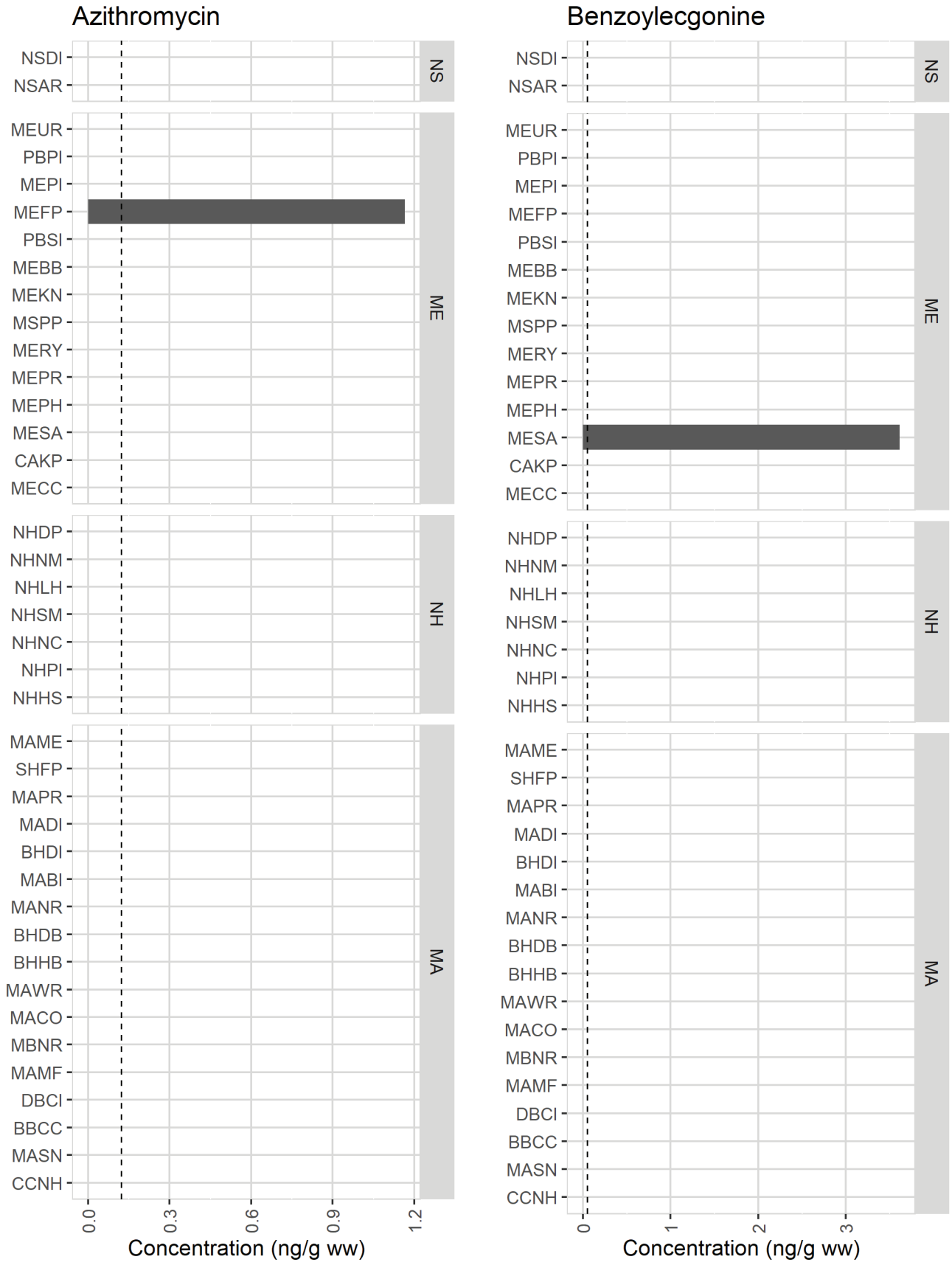

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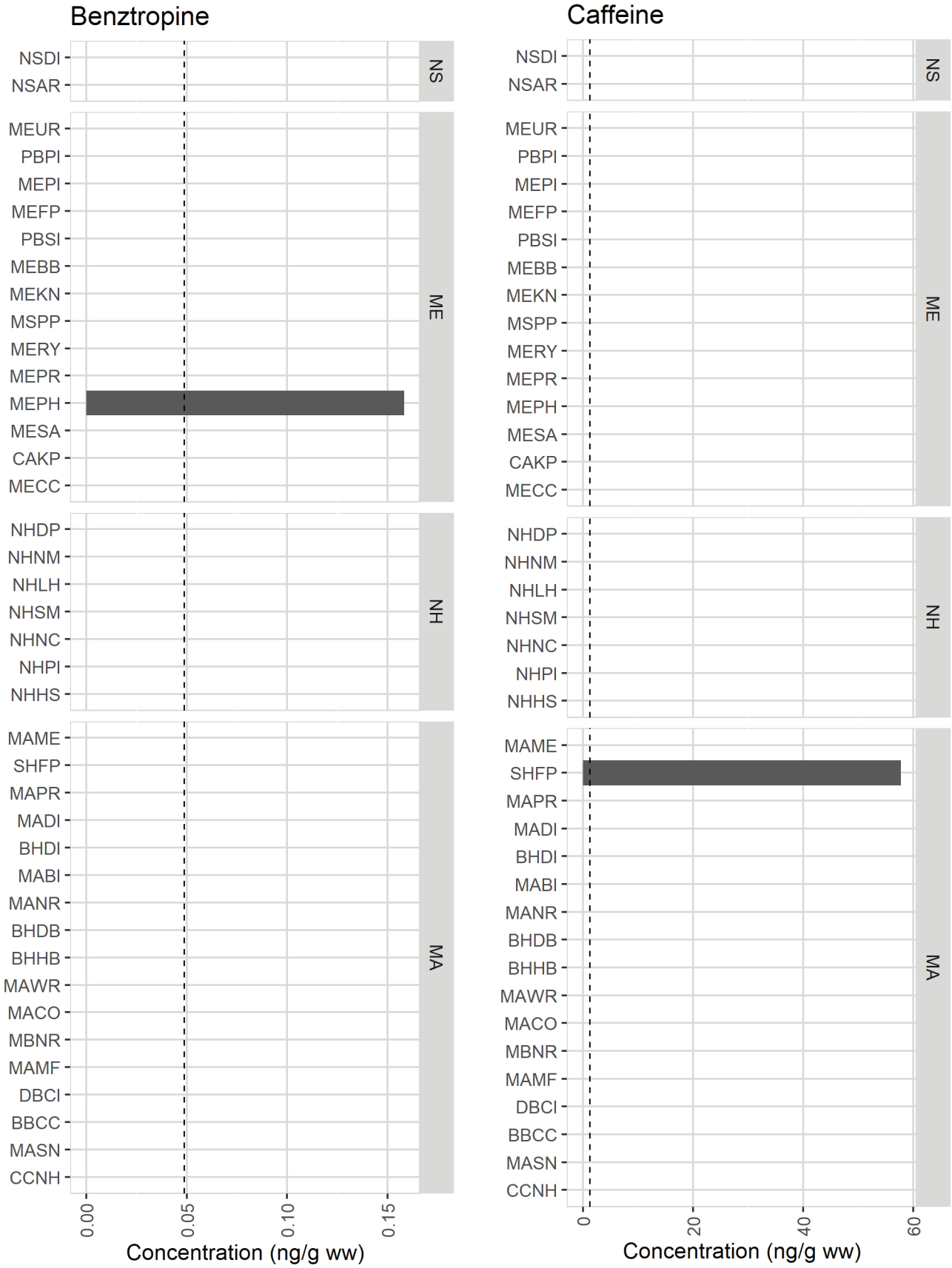
GULF-WIDE ASSESSMENT



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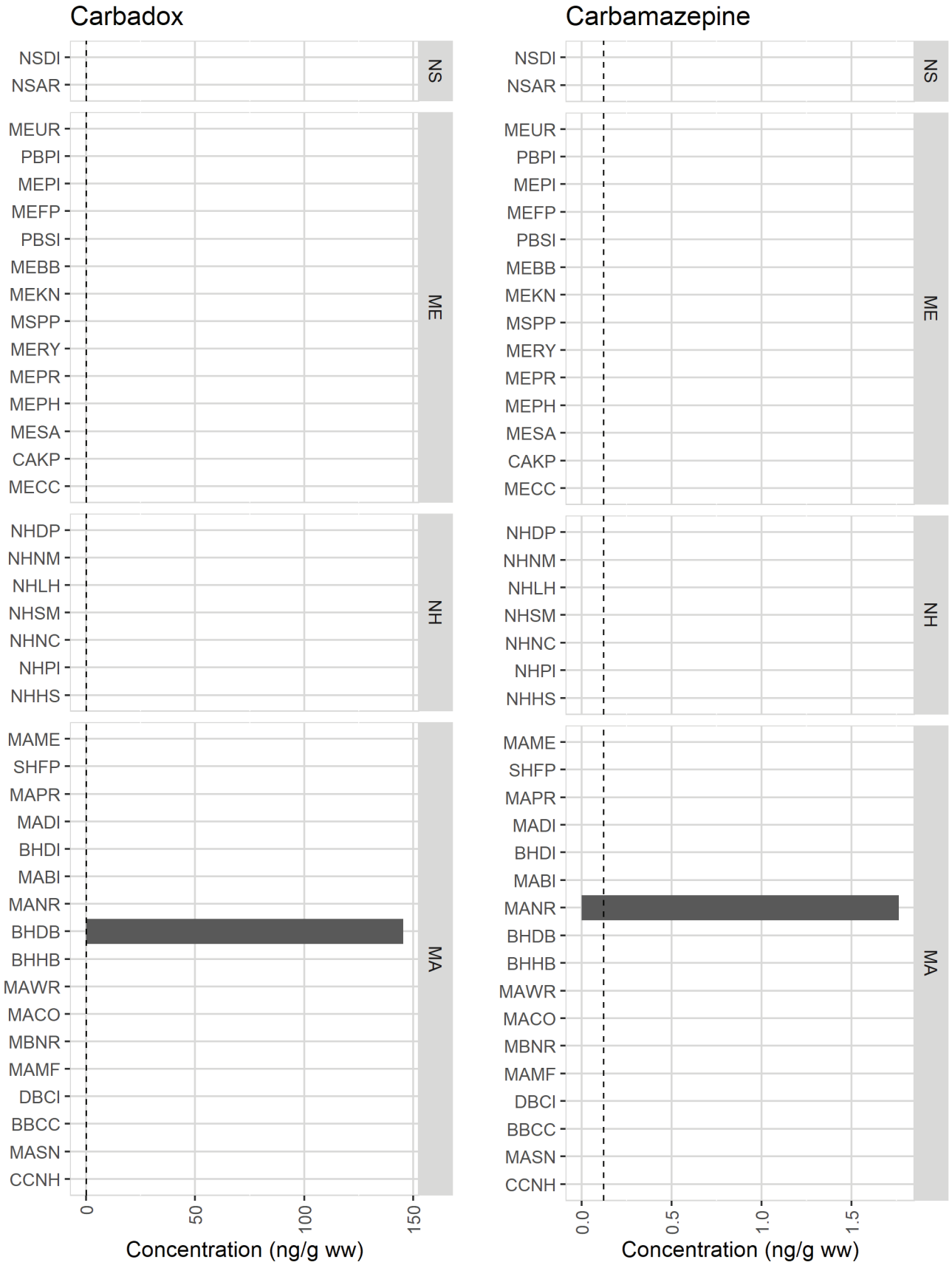

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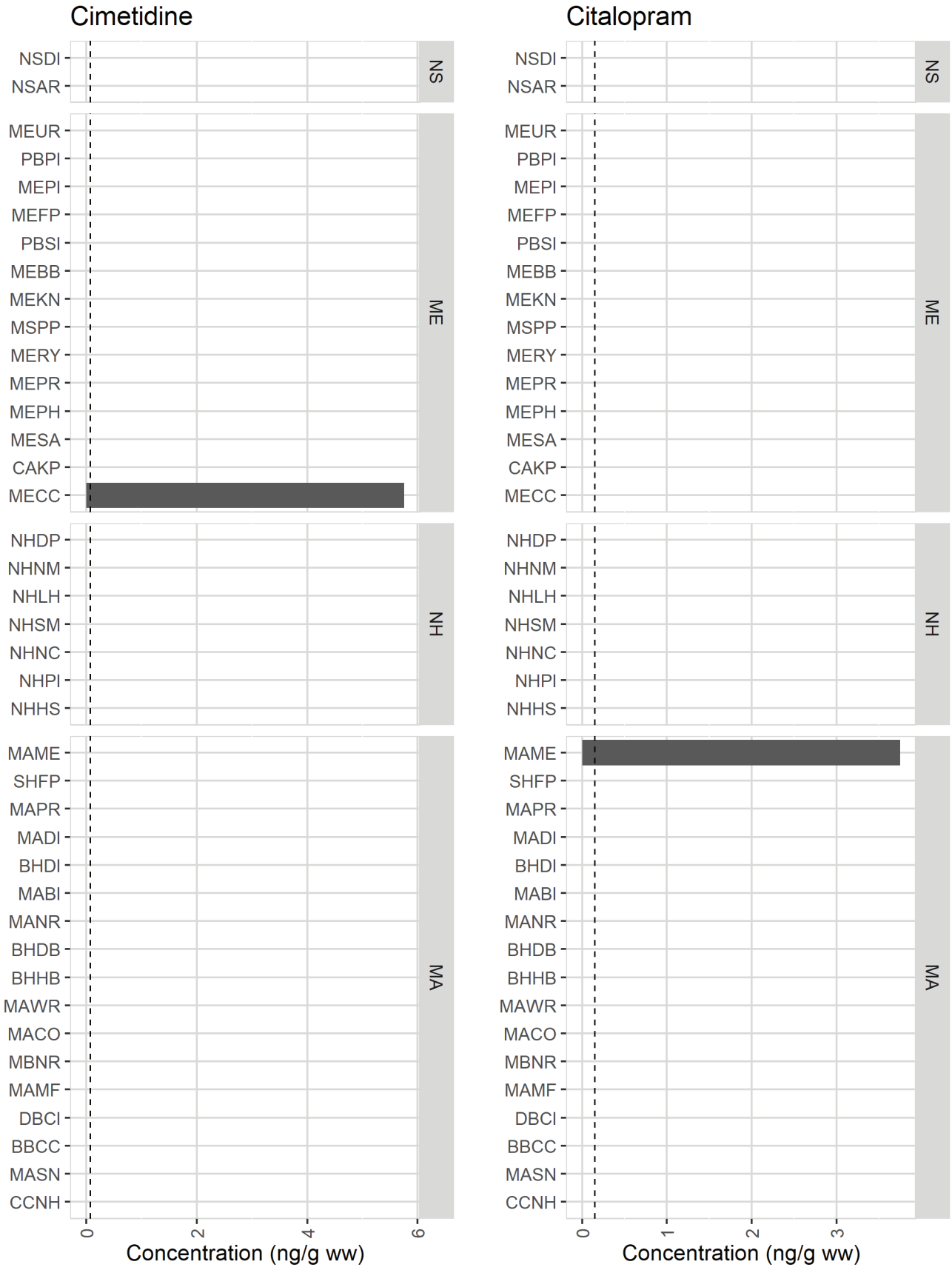

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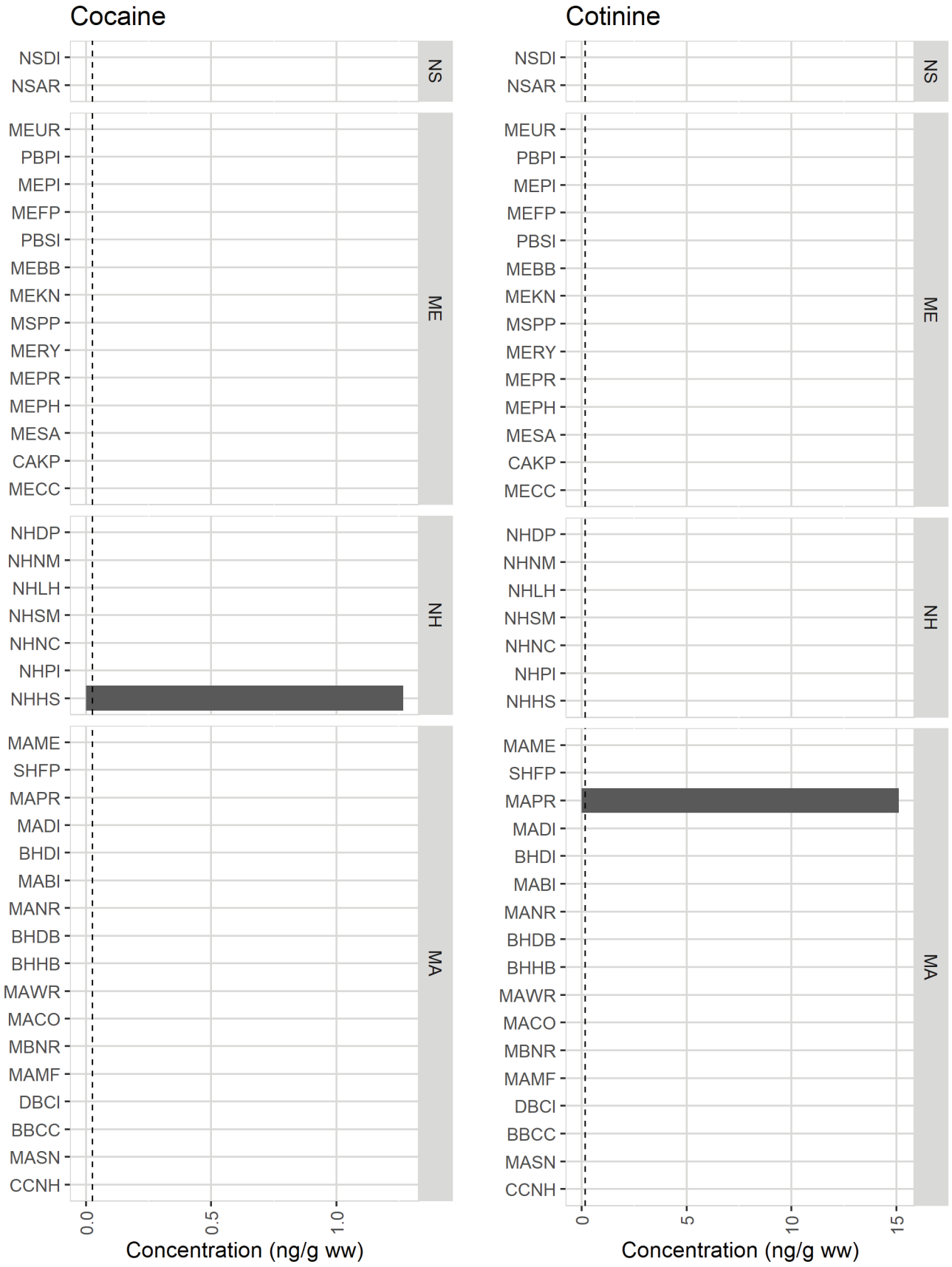

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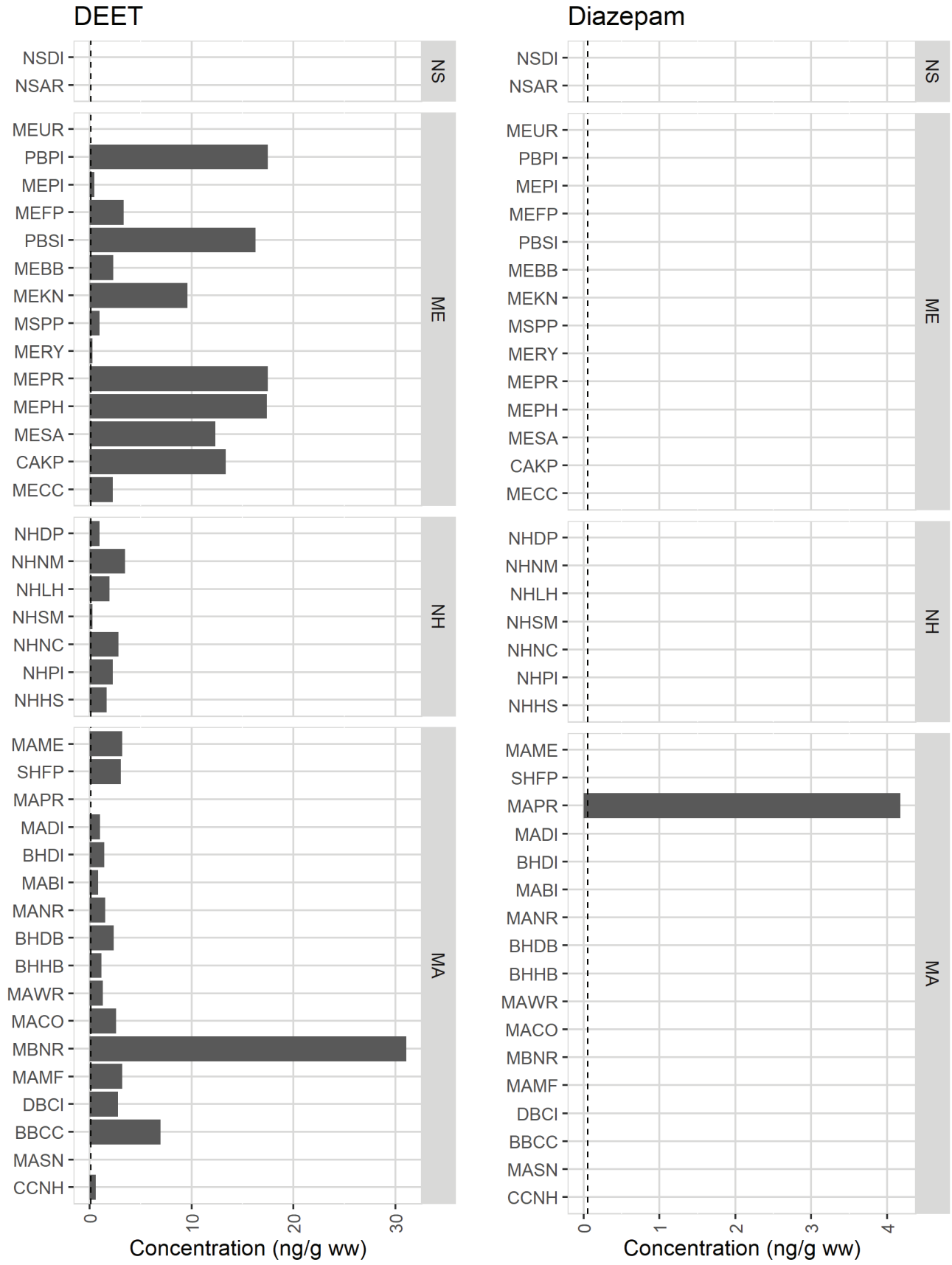
GULF-WIDE ASSESSMENT



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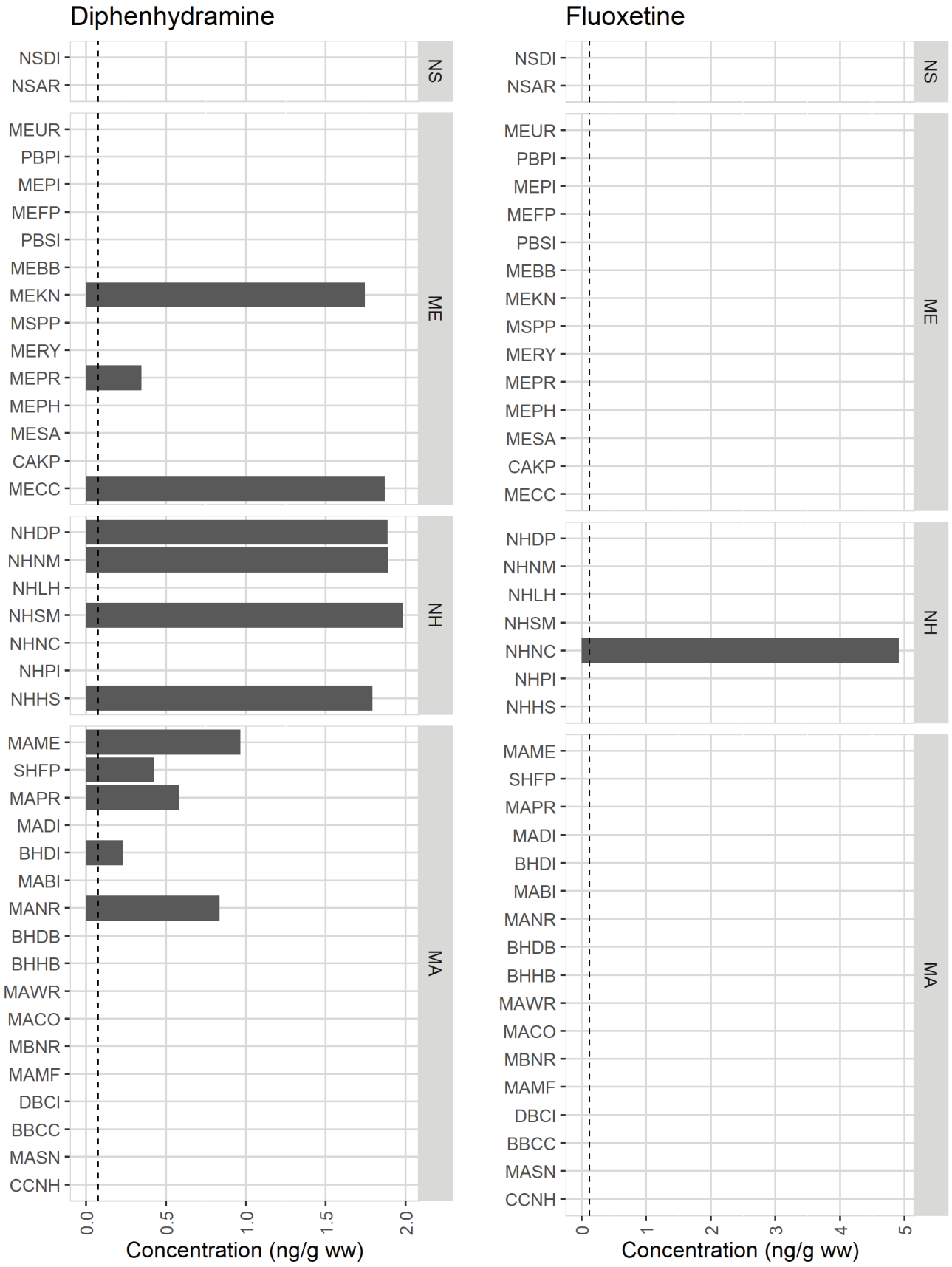
GULF-WIDE ASSESSMENT



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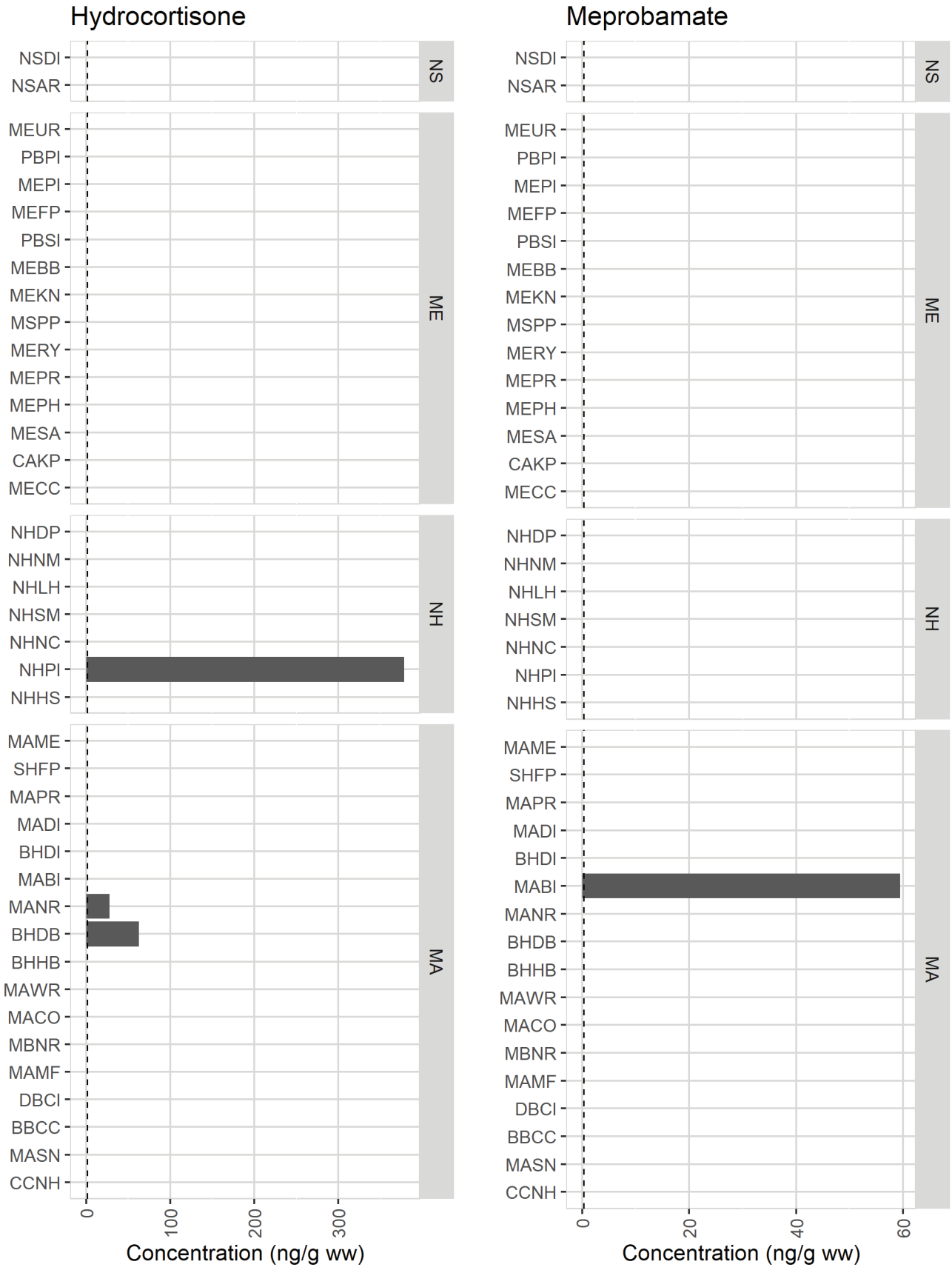

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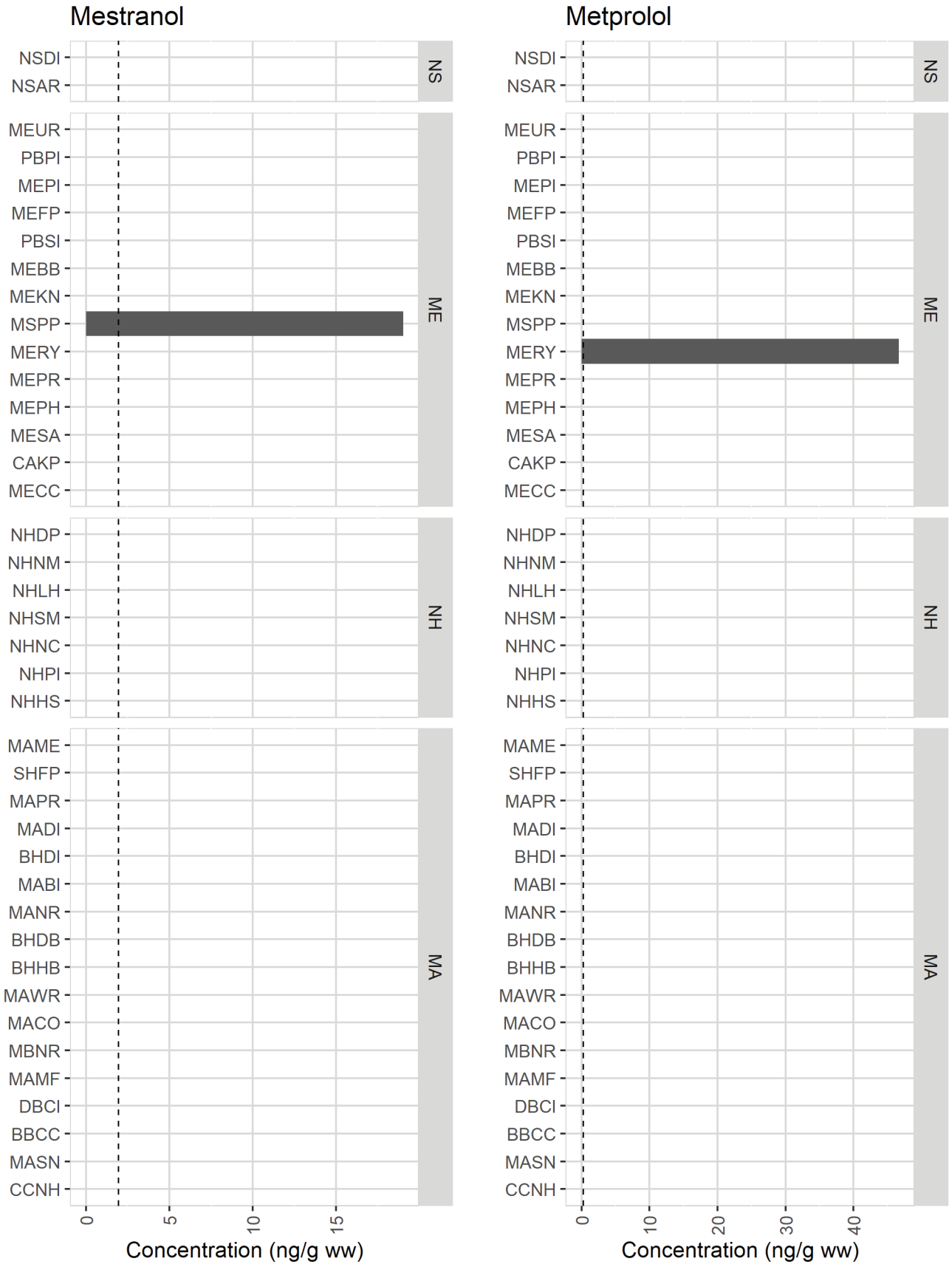

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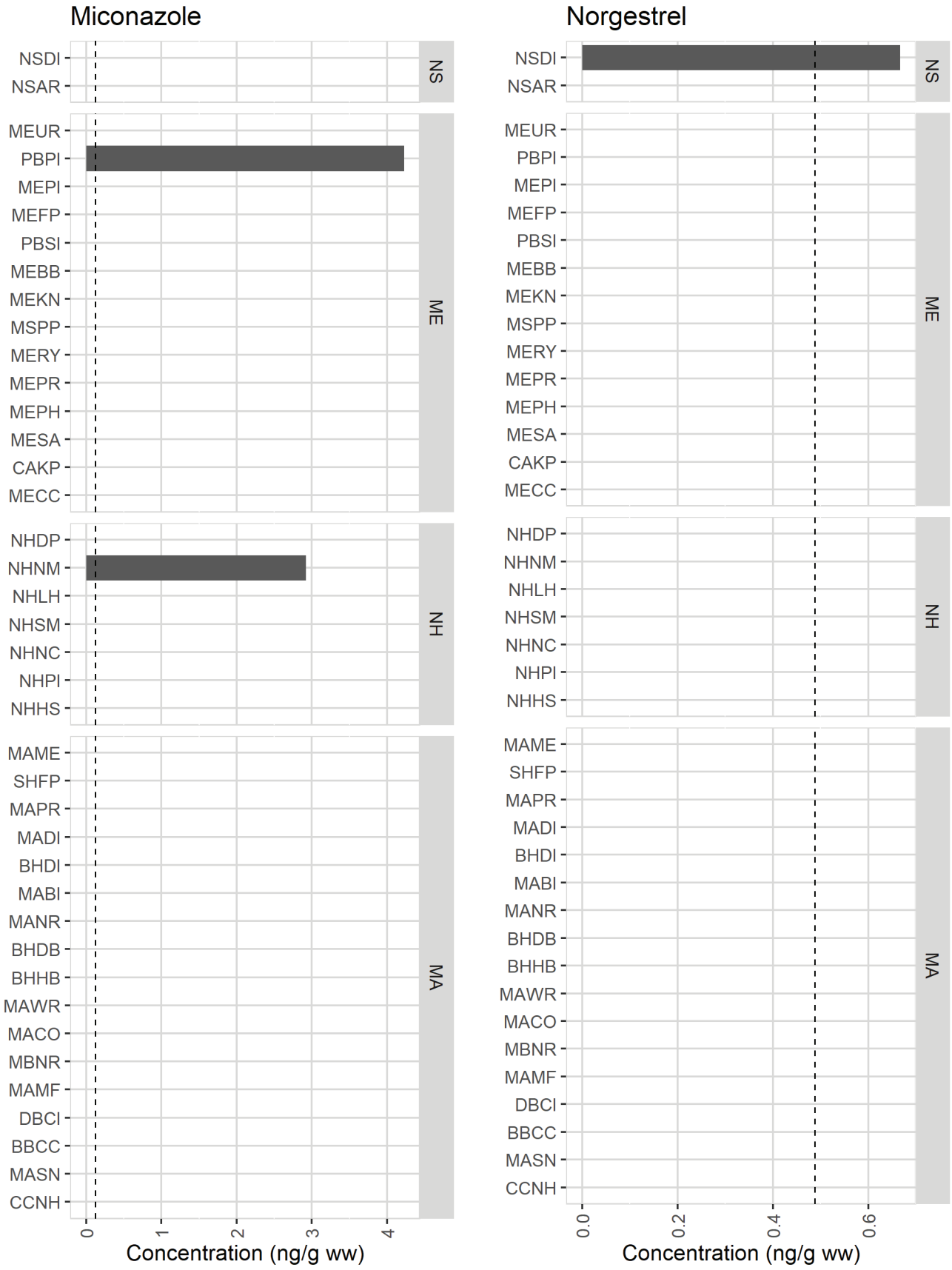

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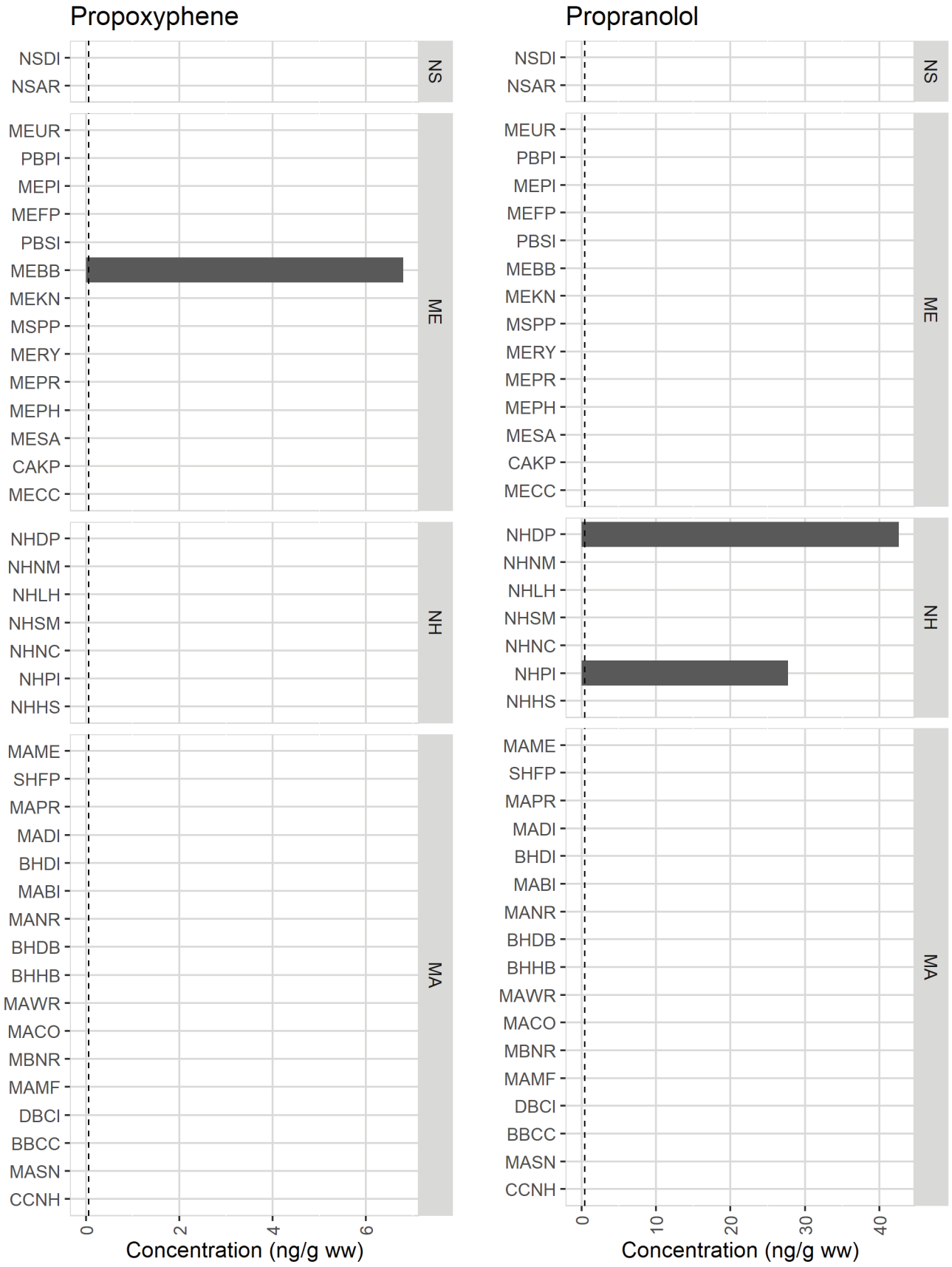

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GULF-WIDE ASSESSMENT

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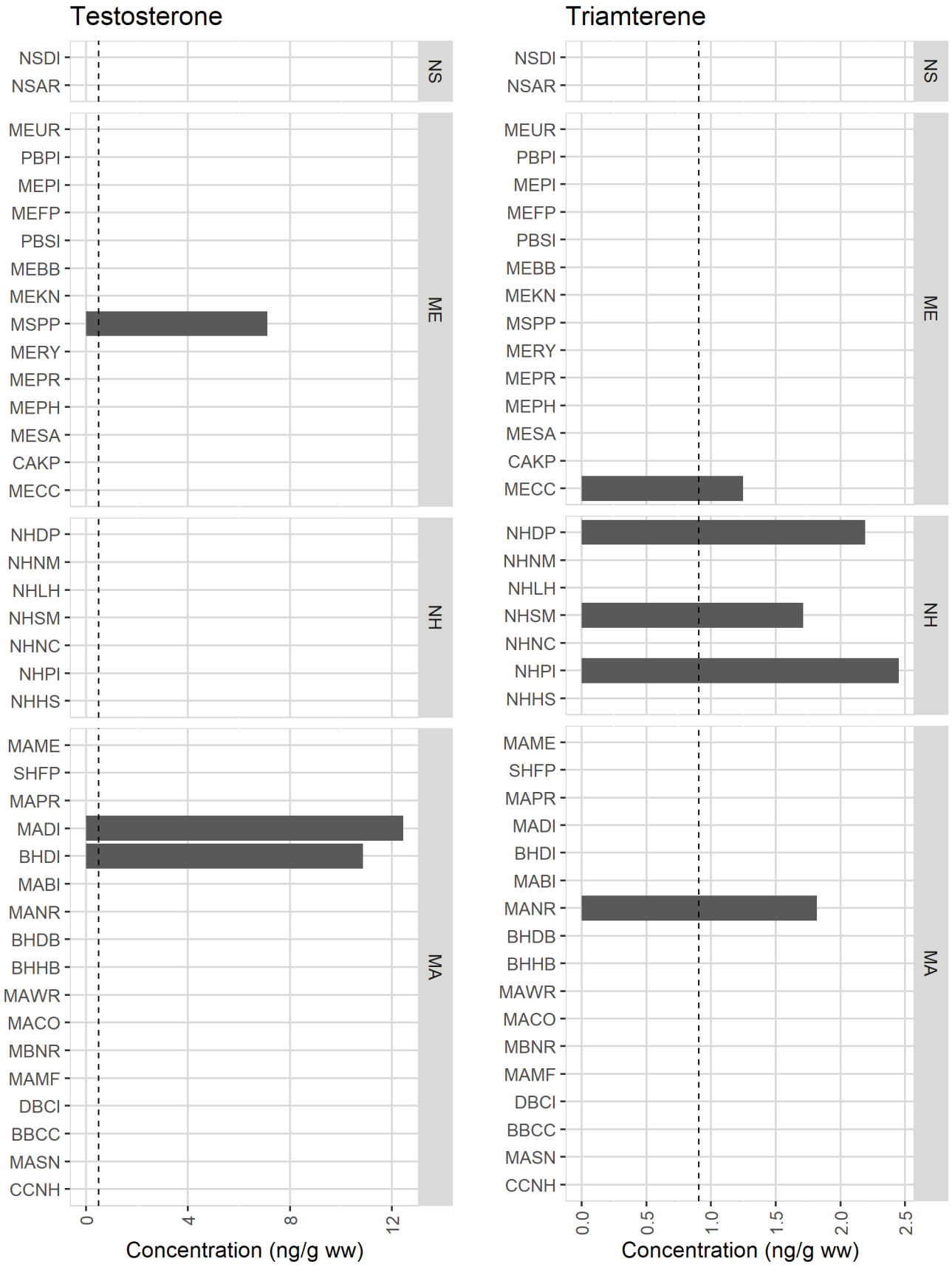

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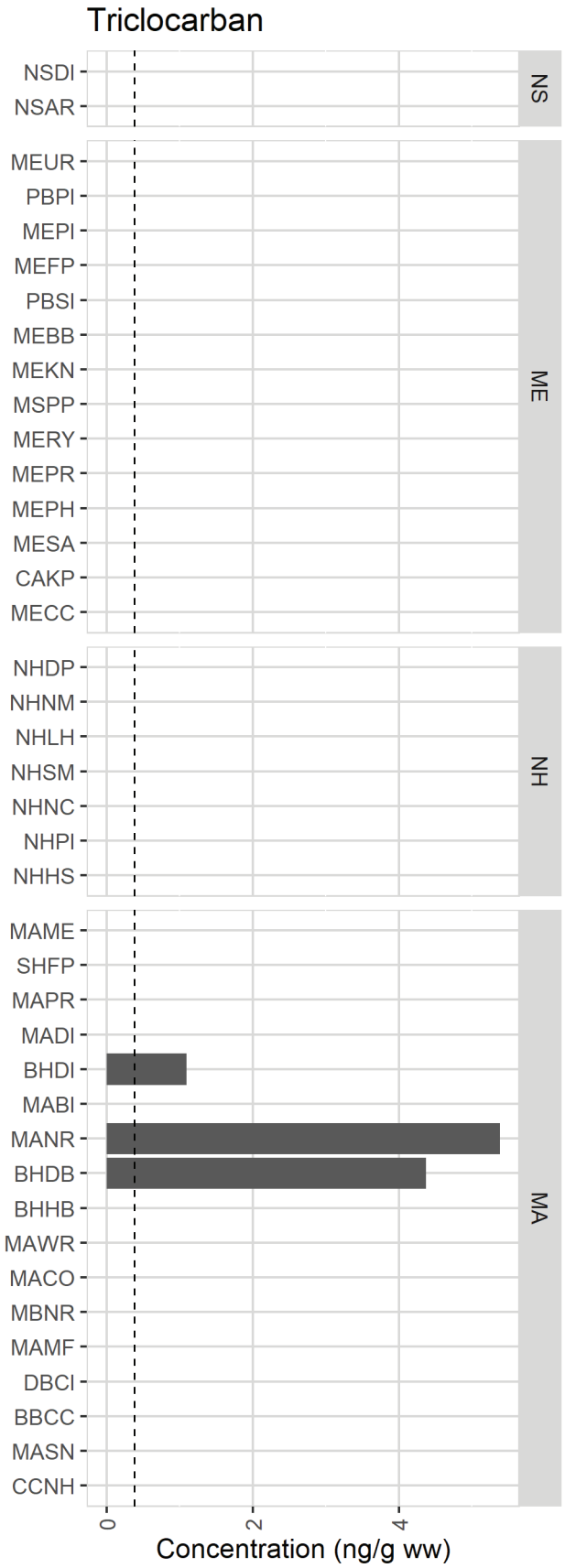
GULF-WIDE ASSESSMENT



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Summary of PPCPs in mussel tissue

- Due to insufficient sample mass at site NSFI, PPCP contaminants were measured in 40 mussel tissue samples (17 in MA, 14 in ME, 7 in NH and 2 in NS).
- 31 of the 121 PPCP contaminants were detected across the study area including the Nova Scotia jurisdiction in Canada, where only residue of norgestrel, a birth control medication, was detected at one site (Figure 23a-g).
- The most frequently detected PPCPs in the Gulf of Maine were DEET, an insect repellant with a Gulf-wide detection frequency of 87.5% (35 sites), followed by sertraline, an antidepressant drug (42.5% - 17 sites), diphenhydramine, an antihistamine drug (30% - 12 sites), ranitidine, a gastroesophageal and heartburn drug (22.5% - 9 sites), and triamterene, a diuretic drug (12.5% - 5 sites) (Table 24).
- Hydrocortisone, a skin condition drug, testosterone, and triclocarban, an antibacterial chemical, were found at three sites each in MA and NH, MA and ME, and only MA respectively.
- 17β-estradiol, a steroidal estrogen, miconazole, an antifungal medication, and propranolol, a hypertension and heart rhythm disorder drug, were detected at two sites each Gulf-wide (Table 24, Figure 23a-g).
- The remainder of the detected PPCP contaminants were found at a single site throughout the Gulf (Table 24).
- The concentration of PPCP contaminants detected varied greatly in mussel tissue across the Gulf (Figure 24).
- Two of the highest concentration values of 378 and 63.0 ng/g ww were recorded for hydrocortisone at the NHPI and BHDB sites in the NH jurisdiction respectively (Figure 24, Appendix 4).
- Carbadox, an antibiotic, was only detected at BHDB in the MA jurisdiction but had a concentration of 145 ng/g ww (Figure 24, Appendix 4).
- Meprobamate, a sedative drug used for insomnia and psychiatric anxiety, and caffeine were both not frequently detected, however, they were found at concentrations of 59.4 and 57.7 ng/g ww respectively at the MABI and SHFP sites in MA (Appendix 4).
- Metprolol and propranolol, which are both used to treat angina and hypertension, were detected respectively at 46.7 and 42.6 ng/g ww in mussel tissues from the sites MERY in ME and NHDP in NH (Appendix 4).
- PPCP contaminants were indiscriminately found in every land-use category in the Gulf of Maine. Contaminants such as DEET, the active ingredient in insect repellent products was found throughout the study area in developed harbor locations, undeveloped areas, and even at open-water sites (Table 4). The detection of PPCP contaminants in all types of land-use category attest to the ubiquity nature of the contaminants in coastal environments.
- PPCP detection frequencies, sertraline concentrations and diphenhydramine concentrations were all positively correlated with percent impervious surface in the 1, 2, 3, 4 and 5 km buffers (Appendix 6). PPCP detection frequencies at sites in the developed land-use category were higher than sites in the undeveloped land-use category in the 4 km buffers ($p=.047$) and were higher than sites in both the undeveloped and open-water land-use categories in the 1 km buffer ($p=.015$). Diphenhydramine concentrations were higher at sites in the developed land-use category than the undeveloped and open-water land-use categories in the 1 km buffer ($p=.032$). There was no correlation between DEET concentrations and percent impervious surface or land-use categories.

Coast of Maine. Credit: NOAA

Jurisdiction-specific assessment: MASSACHUSETTS SUMMARY

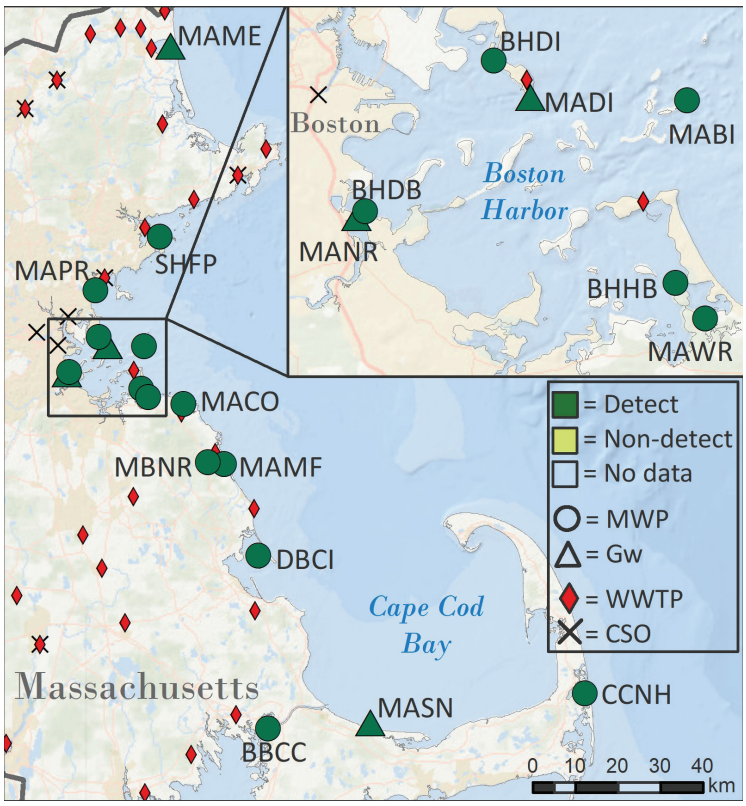

Figure 25. Map of MA jurisdiction highlighting location of sites with PPCP detection in mussel tissue.

Table 26. PPCP compounds frequency of detection in mussel tissue from MA jurisdiction (n = 17).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
DEET	15	17	88.2
Sertraline	6	17	35.3
Diphenhydramine	5	17	29.4
Ranitidine	3	17	17.6
Triclocarban	3	17	17.6
Hydrocortisone	2	17	11.8
Testosterone	2	17	11.8
Androstenedione	1	17	5.9
Caffeine	1	17	5.9
Carbadox	1	17	5.9
Carbamazepine	1	17	5.9
Citalopram	1	17	5.9
Cotinine	1	17	5.9
Diazepam	1	17	5.9
Meprobamate	1	17	5.9
Triamterene	1	17	5.9

Summary of PPCPs in Massachusetts

- Mussel tissue samples from a total of 17 sites were tested for PPCP contaminants in MA (Figure 25).
- One or more PPCP contaminants were detected at every monitoring site in MA (Figure 25).
- The most frequently detected PPCP contaminants in MA were DEET (88.2%), sertraline (35.3%), diphenhydramine (29.4%), ranitidine (17.6%), triclocarban (17.6%), hydrocortisone (11.8%) and testosterone (11.8%) (Table 26).
- The remainder of the detected PPCP contaminants were found at single sites throughout the MA jurisdiction (Table 26).
- The concentration of PPCP contaminants detected varied from 0.23 ng/g ww for diphenhydramine, an allergy antihistamine product at the Boston Harbor Deer Island (BHDI) site, to 145 ng/g ww for carbadox, an animal antibiotic at the Boston Harbor Dochester Bay (BHDB) site (Figure 24, Appendix 4). Concentrations of 63.0 ng/g ww of hydrocortisone, a skin condition medication, 59.4 ng/g ww of meprobamate, an anxiety treatment medication, and 57.7 ng/g ww of caffeine were found respectively at the Boston Harbor Dochester Bay (BHDB), Boston Harbor Brewster Island (MABI) and at the Salem Harbor Folger Point (SHFP) sites in MA.
- PPCP contaminants were present at all monitoring sites in the state. While PPCPs were found in all land-use categories, they were more frequently found in the developed Boston Harbor area than in other coastal locations in the study area. The higher detection rates and concentrations of PPCP contaminants in the Boston Harbor area might be linked to the wastewater treatment plants and combines sewer outfalls in this area (Figure 25).

Jurisdiction-specific assessment: NEW HAMPSHIRE SUMMARY

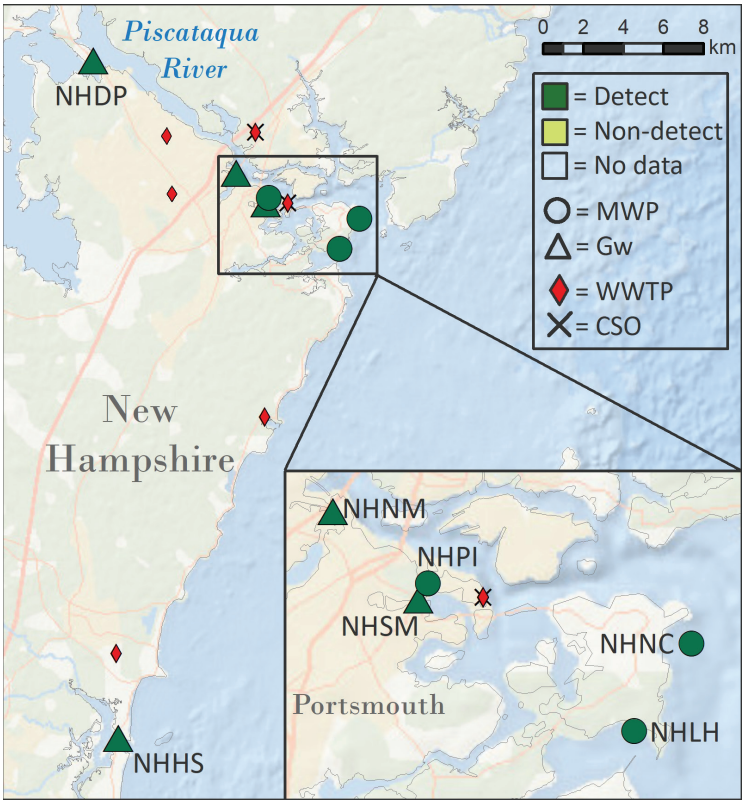

Figure 26. Map of NH jurisdiction highlighting location of sites with PPCP detection in mussel tissue.

Table 27. PPCP compounds frequency of detection in mussel tissue from NH jurisdiction (n = 7).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
DEET	7	7	100.0
Sertraline	5	7	71.4
Diphenhydramine	4	7	57.1
Triamterene	3	7	42.9
Propranolol	2	7	28.6
Amitriptyline	1	7	14.3
Atenolol	1	7	14.3
Cocaine	1	7	14.3
Fluoxetine	1	7	14.3
Hydrocortisone	1	7	14.3
Miconazole	1	7	14.3

Summary of PPCPs in New Hampshire

- Mussel tissue samples from a total of seven sites were tested for PPCP contaminants in NH (Figure 26).
- At least one PPCP contaminant was detected at every monitoring site in NH (Figure 26).
- The most frequently detected PPCP contaminants in NH were DEET, the active ingredient in insect repellents (100%), sertraline, an antidepressant drug (71.4%), diphenhydramine, an allergy antihistamine product (57.1%), triamterene, a diuretic pill (42.9%), and propranolol, a hypertension medication (28.6%) (Table 27).
- The remainder of the detected PPCP contaminants were found at single site throughout the NH study area (Table 27).
- The concentration of PPCP contaminants detected varied from 0.28 ng/g ww for DEET at the South Mill Point (NHSM) site to 378 ng/g ww for hydrocortisone, a skin condition medication, at the Pierce Island (NHPI) site (Figure 24, Appendix 4). Concentrations 42.6 ng/g ww and 27.7 ng/g ww of propranolol and 4.91 ng/g ww of fluoxetine, an antidepressant medication, were found respectively at the Piscataqua River Dover Point (NHDP), Pierce Island (NHPI) and New Castle (NHNC) sites in NH.
- PPCP contaminants were present at all monitoring sites in the state in developed, undeveloped and open-water land-use categories. The discharges of wastewater treatment plants and combined sewer outfalls within the coastal watersheds may have contributed to the presence and distribution of PPCP in the NH study area (Figure 26).

Jurisdiction-specific assessment: MAINE SUMMARY

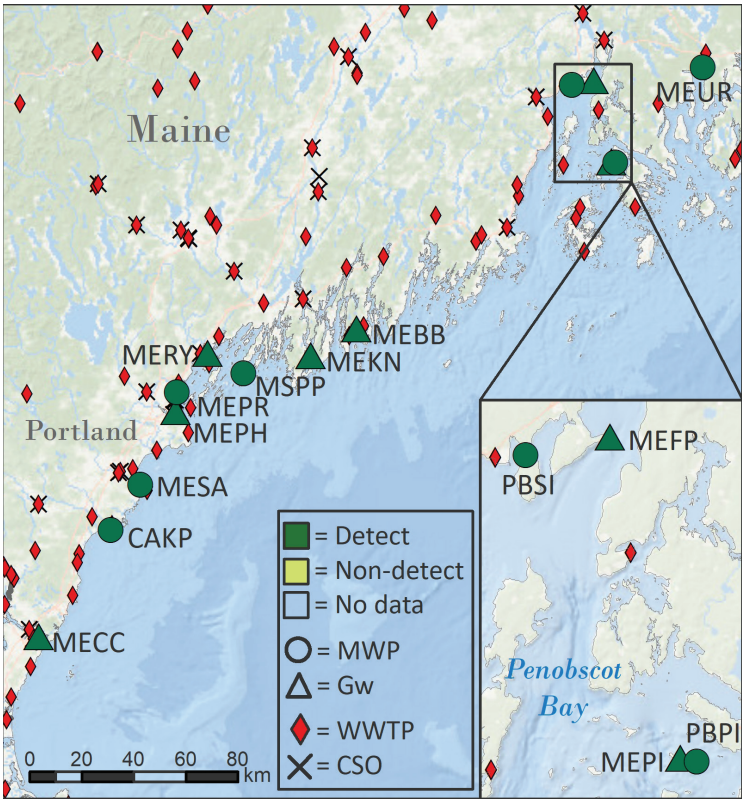

Figure 27. Map of ME jurisdiction highlighting location of sites with PPCP detection in mussel tissue.

Table 28. PPCP compounds frequency of detection in mussel tissue from ME jurisdiction (n = 14).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
DEET	13	14	92.9
Ranitidine	6	14	42.9
Sertraline	6	14	42.9
Diphenhydramine	3	14	21.4
17β-estradiol	2	14	14.3
Azithromycin	1	14	7.1
Benzoylecgonine	1	14	7.1
Benztropine	1	14	7.1
Cimetidine	1	14	7.1
Mestranol	1	14	7.1
Metprolol	1	14	7.1
Miconazole	1	14	7.1
Propoxyphene	1	14	7.1
Testosterone	1	14	7.1
Triamterene	1	14	7.1

Summary of PPCPs in Maine

- Mussel tissue samples from a total of 14 sites were tested for PPCP contaminants in ME (Figure 27).
- One or more PPCP contaminants were detected at each of the 14 sites in ME (Figure 27).
- DEET, the most active ingredient in insect repellents, was the most frequently detected PPCP contaminant in ME (92.9%). Ranitidine, the heartburn medication, and sertraline, an antidepressant drug, were each detected at 42.9%. Diphenhydramine, an antihistamine medication, was found at 21.4% of the sites, and 17β-estradiol, a postmenopausal estrogen replacement therapy drug, was detected a 14.3% of the monitoring sites (Table 28).
- The remainder of the detected PPCP contaminants were found at single sites throughout the ME jurisdiction (Table 28).
- In ME, the concentration of PPCP contaminants detected varied from 0.16 ng/g ww for benztropine, an anticholinergic drug used to improve mobility in Parkinsons disease, at the Stroudwater-Force Portland Harbor (MEPH) site to 46.7 ng/g ww for metprolol, a hypertension medication, at the Royal River (MERY) site (Appendix 4). The highest concentrations of DEET in ME (17.5, 17.5, 17.4, 16.3, 13.4, 12.3 and 9.60 ng/g ww) were found at Presumpscot River (MEPR), Penobscot Bay Pierces Island (PBPI), Stroudwater-Force Portland Harbor (MEPH), Penobscot Bay Sears Island (PBSI), Cape Arundel Kennebunkport (CAKP), Saco River (MESA), and Kennebec Perkins Island (MEKN) (respectively). The concentrations of 19.0 ng/g ww, for the birth control drug mestranol and 7.11 ng/g ww, for testosterone hormone, were measured in mussels from the Merriconeag Sound Potts Point (MSPP). Ranitidine, used for heartburn, was found at 16.8

Summary of PPCPs in Maine (cont.)

ng/g ww at the Penobscot Bay Pickering Island (PBPI), the antidepressant sertraline was detected at 14.2 ng/g ww at the Stroudwater-Fore Portland Harbor (MEPH), and propoxyphene, a narcotic pain-reliever, was measured at a concentration of 6.80 ng/g ww at the Boothbay Harbor (MEBB) site (Figure 24, Appendix 4)

- PPCP contaminants present at all monitoring sites in the state in developed, undeveloped and open-water land-use categories. Discharges from wastewater treatment plants and combined sewer outfalls within the coastal watersheds might be the most important source of the presence and wide distribution of PPCP contaminants in the ME coastal monitoring area.

Jurisdiction-specific assessment: Nova Scotia Summary

Figure 28. Map of NS jurisdiction highlighting location of sites with PPCP detection in mussel tissue.

Summary of PPCPs in Nova Scotia

- Mussel tissue samples from two monitoring sites in NS were measured for PPCP contaminants (Figure 28).
- The only PPCP contaminant recorded in the NS jurisdiction was norgestrel at the Annapolis Basin Digby (NSDI) site in Canada (Figure 28, Table 29).
- Norgestrel is a hormone that is used to prevent pregnancy and it was found at a concentration 0.67 ng/g ww (Appendix 4).

Table 29. PPCP compounds frequency of detection in mussel tissue from NS jurisdiction (n = 2).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
Norgestrel	1	2	50.0

Brominated Flame Retardants (BFRs)

Chemical Description

Brominated flame retardants (BFRs), such as polybrominated diphenyl ethers (PBDEs) and polybrominated biphenyls (PBBs), are a group of chemicals with 209 possible unique congeners that are used in firefighting materials and in consumer and household products to reduce flammability.

Commercially, three types of PBDE industrial mixtures have been available, the pentabromodiphenyl ether (penta-BDE), octabromodiphenyl ether (octa-BDE) and the decabromodiphenyl ether (deca-BDE) mixtures (EPA, 2014b). As these products age and degrade, PBDEs leach into the environment. PBDEs have become ubiquitous in the environment and are detected in materials that include household dust, human breast milk, sediment and wildlife (ATSDR, 2015). The less brominated PBDEs, like tetra-, penta- and hexa-BDE, demonstrate high affinity for lipids and tend to bioaccumulate in animals and humans, while highly brominated PBDEs like deca-BDE tend to absorb more onto sediment and soil. The toxicology of PBDEs is not well understood, but PBDEs have been associated with tumors, neurodevelopmental toxicity and thyroid hormone imbalance. Some PBDE congeners have hepatotoxic and mutagen effects while others may act as estrogen receptor agonists in vitro (Meerts et al., 2001). Due to ubiquitous distribution, persistence and potential for toxicity, the manufacturing of the 'penta' and 'octa' PBDEs mixtures have been phased out starting in 2004, and the deca mixture starting in 2013 (EPA, 2014b; Schreder and La Guardia, 2014).

PBBs are manufactured chemicals primarily used in firefighting materials. Like PBDEs, PBBs are classified as persistent organic pollutants (POPs), however, their environment impacts are not well understood. Although it is not definitively known whether PBBs can cause cancer in human beings, cancer in lab mice exposed to very high concentrations has been observed. As a result of these animal tests, the United States Department of Health and Human Services has concluded that PBBs might reasonably be characterized as carcinogens (Wang, 2009). The application of PBB in firefighting materials is now controlled as a hazardous substance (Safe, 1984).

The list of BFRs contaminants measured in this study is presented in Table 30a-b. In this study, the BFR analyses were performed by TDI-Brooks International Inc. following procedures used by the NOAA NS&T Program (Kimbrough et al., 2007).

NOTE: Since none of the 19 PBB compounds were detected in any of the 41 samples Gulf-wide in this study, they have been excluded from the data analysis and only PDBEs are reported.

Chemical Description

Table 30a. PBB compounds tested (n=19).

Chemical code	Chemical name
PBB 1	PBB 1 (2-MonoBB)
PBB 2	PBB 2 (3-MonoBB)
PBB 3	PBB 3 (4-MonoBB)
PBB 4	PBB 4 (2,2'-DiBB)
PBB 7	PBB 7 (2,4-DiBB)
PBB 9	PBB 9 (2,5-DiBB)
PBB 10	PBB 10 (2,6-DiBB)
PBB 15	PBB 15 (4,4'-DiBB)
PBB 18	PBB 18 (2,2',5-TriBB)
PBB 26	PBB 26 (2,3',5-TriBB)
PBB 30	PBB 30 (2,4,6-TriBB)
PBB 31	PBB 31 (2,4',5-TriBB)
PBB 49	PBB 49 (2,2',4,5'-TetraBB)
PBB 52	PBB 52 (2,2',5,5'-TetraBB)
PBB 53	PBB 53 (2,2',5,6'-TetraBB)
PBB 77	PBB 77 (3,3',4,4'-TetraBB)
PBB 80	PBB 80 (3,3',5,5'-TetraBB)
PBB 103	PBB 103 (2,2',4,5',6-PentaBB)
PBB 155	PBB 155 (2,2',4,4',6,6'-HexaBB)

Table 30b. PBDE compounds tested (n=51).

Chemical code	Chemical name
PBDE-1	BDE 1 (2-MonoBDE)
PBDE-2	BDE 2 (3-MonoBDE)
PBDE-3	BDE 3 (4-MonoBDE)
PBDE-7	BDE 7 (2,4-DiBDE)
PBDE-8	BDE 8 (2,4'-DiBDE)
PBDE-10	BDE 10 (2,6-DiBDE)
PBDE-11	BDE 11 (3,3'-DiBDE)
PBDE-12	BDE 12 (3,4-DiBDE)
PBDE-13	BDE 13 (3,4'-DiBDE)
PBDE-15	BDE 15 (4,4'-DiBDE)
PBDE-17	BDE 17 (2,2',4-TriBDE)
PBDE-25	BDE 25 (2,3',4-TriBDE)
PBDE-28	BDE 28 (2,4,4'-TriBDE)
PBDE-30	BDE 30 (2,4,6-TriBDE)

Table 30b (cont). PBDE compounds tested (n=51).

Chemical code	Chemical name
PBDE-32	BDE 32 (2,4',6-TriBDE)
PBDE-33	BDE 33 (2',3,4-TriBDE)
PBDE-35	BDE 35 (3,3',4-TriBDE)
PBDE-37	BDE 37 (3,4,4'-TriBDE)
PBDE-47	BDE 47 (2,2',4,4'-TetraBDE)
PBDE-66	BDE 66 (2,3',4,4'-TetraBDE)
PBDE-71/49	BDE 49/71 (2,2',4,5'-TetraBDE/2,3',4',6-TetraPDE)
PBDE-75	BDE 75 (2,4,4',6-TetraBDE)
PBDE-77	BDE 77 (3,3',4,4'-TetraBDE)
PBDE-85	BDE 85 (2,2',3,4,4'-PentaBDE)
PBDE-99	BDE 99 (2,2',4,4',5-PentaBDE)
PBDE-100	BDE 100 (2,2',4,4',6-PentaBDE)
PBDE-116	BDE 116 (2,3,4,5,6-PentaBDE)
PBDE-118	BDE 118 (2,3',4,4',5-PentaBDE)
PBDE-119	BDE 119 (2,3',4,4',6-PentaBDE)
PBDE-126	BDE 126 (3,3',4,4',5-PentaBDE)
PBDE-138	BDE 138 (2,2',3,4,4',5'-HexaBDE)
PBDE-153	BDE 153 (2,2',4,4',5,5'-HexaBDE)
PBDE-154	BDE 154 (2,2',4,4',5,6'-HexaBDE)
PBDE-155	BDE 155 (2,2',4,4',6,6'-HexaBDE)
PBDE-166	BDE 166 (2,3,4,4',5,6-HexaBDE)
PBDE-181	BDE 181 (2,2',3,4,4',5,6-HeptaBDE)
PBDE-183	BDE 183 (2,2',3,4,4',5',6-HeptaBDE)
PBDE-190	BDE 190 (2,3,3',4,4',5,6-HeptaBDE)
PBDE-194	BDE 194 (2,2',3,3',4,4',5,5'-OctaBDE)
PBDE-195	BDE 195 (2,2',3,3',4,4',5,6-OctaBDE)
PBDE-196	BDE 196 (2,2',3,3',4,4',5,6'-OctaBDE)
PBDE-197	BDE 197 (2,2',3,3',4,4',6,6'-OctaBDE)
PBDE-198/199/203/200	BDE 198/199/203/200 (OctaBDE)
PBDE-201	BDE 201 (2,2',3,3',4,5',6,6'-OctaBDE)
PBDE-202	BDE 202 (2,2',3,3',5,5',6,6'-OctaBDE)
PBDE-204	BDE 204 (2,2',3,4,4',5,6,6'-OctaBDE)
PBDE-205	BDE 205 (2,3,3',4,4',5,5',6-OctaBDE)
PBDE-206	BDE 206 (2,2',3,3',4,4',5,5',6-NonaBDE)
PBDE-207	BDE 207 (2,2',3,3',4,4',5,6,6'-NonaBDE)
PBDE-208	BDE 208 (2,2',3,3',4,5,5',6,6-NonaBDE)
PBDE-209	BDE 209 (2,2',3,3',4,4',5,5',6,6'-DecaBDE)

Presence and distribution of PBDEs in mussel tissue: GULF-WIDE ASSESSMENT

Table 31. PBDE compounds Gulf-wide frequency of detection in mussel tissue.

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PBDE-47	33	41	80.5
PBDE-99	26	41	63.4
PBDE-71/49	24	41	58.5
PBDE-119	22	41	53.7
PBDE-77	20	41	48.8
PBDE-100	8	41	19.5
PBDE-126	6	41	14.6
PBDE-66	5	41	12.2
PBDE-209	2	41	4.9
PBDE-118	2	41	4.9
PBDE-197	1	41	2.4
PBDE-1	1	41	2.4
Compound Class Total	150	2098	7.1

Number of compounds detected:
12/51

Number of sites with detects:
37/41

Most detected compound:
PBDE-47

Table 32. PBDE compounds number of detects in mussel tissue at each site.

Site	State	Number of Detects	Number of Compounds Analyzed
MAME	MA	9	51
MANR	MA	7	51
DBCI	MA	6	51
MAWR	MA	6	51
NHHS	NH	6	51
BHDB	MA	5	51
MABI	MA	5	51
MAPR	MA	5	51
MEBB	ME	5	51
MEFP	ME	5	51
NHNC	NH	5	51
NHNM	NH	5	51
NHDP	NH	5	51
MAMF	MA	4	51
MACO	MA	4	51
BHHB	MA	4	51
MADI	MA	4	51
MECC	ME	4	51
MERY	ME	4	51
MEPI	ME	4	51
NHPI	NH	4	51
NHLH	NH	4	51
NSAR	NS	4	51
NSFI	NS	4	51
MASN	MA	3	51
BHDI	MA	3	51
SHFP	MA	3	51
MESA	ME	3	51
MEPH	ME	3	51
MEPR	ME	3	51
PBSI	ME	3	51
NHSM	NH	3	51
MSPP	ME	2	51
MEKN	ME	2	51
NSDI	NS	2	51
MBNR	MA	1	51
CAKP	ME	1	51

Gulf-wide Assessment

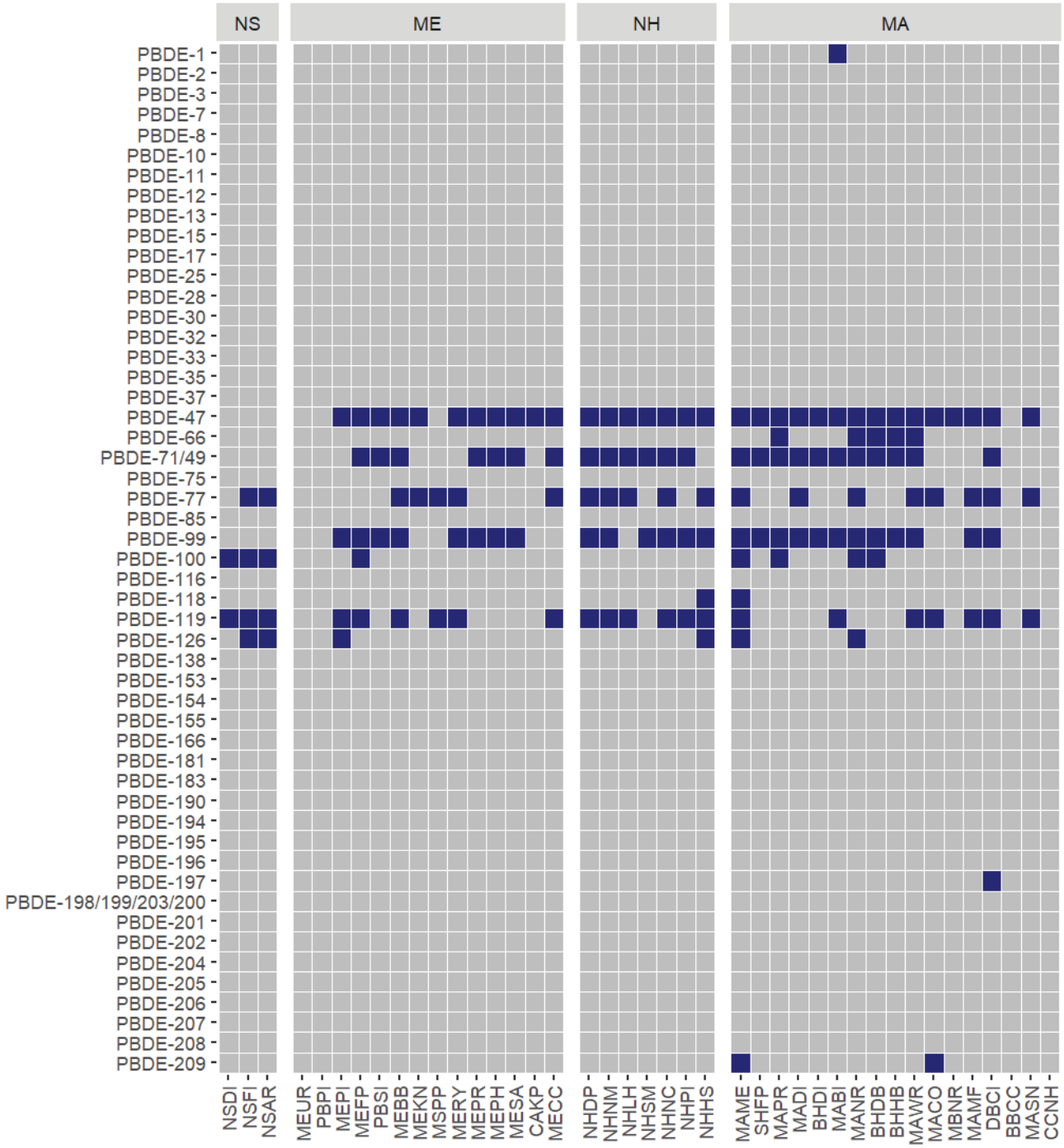

Figure 29. Distribution map showing presence (■) and absence (■) of PBDE compounds measured in mussel tissues from the Gulf of Maine. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

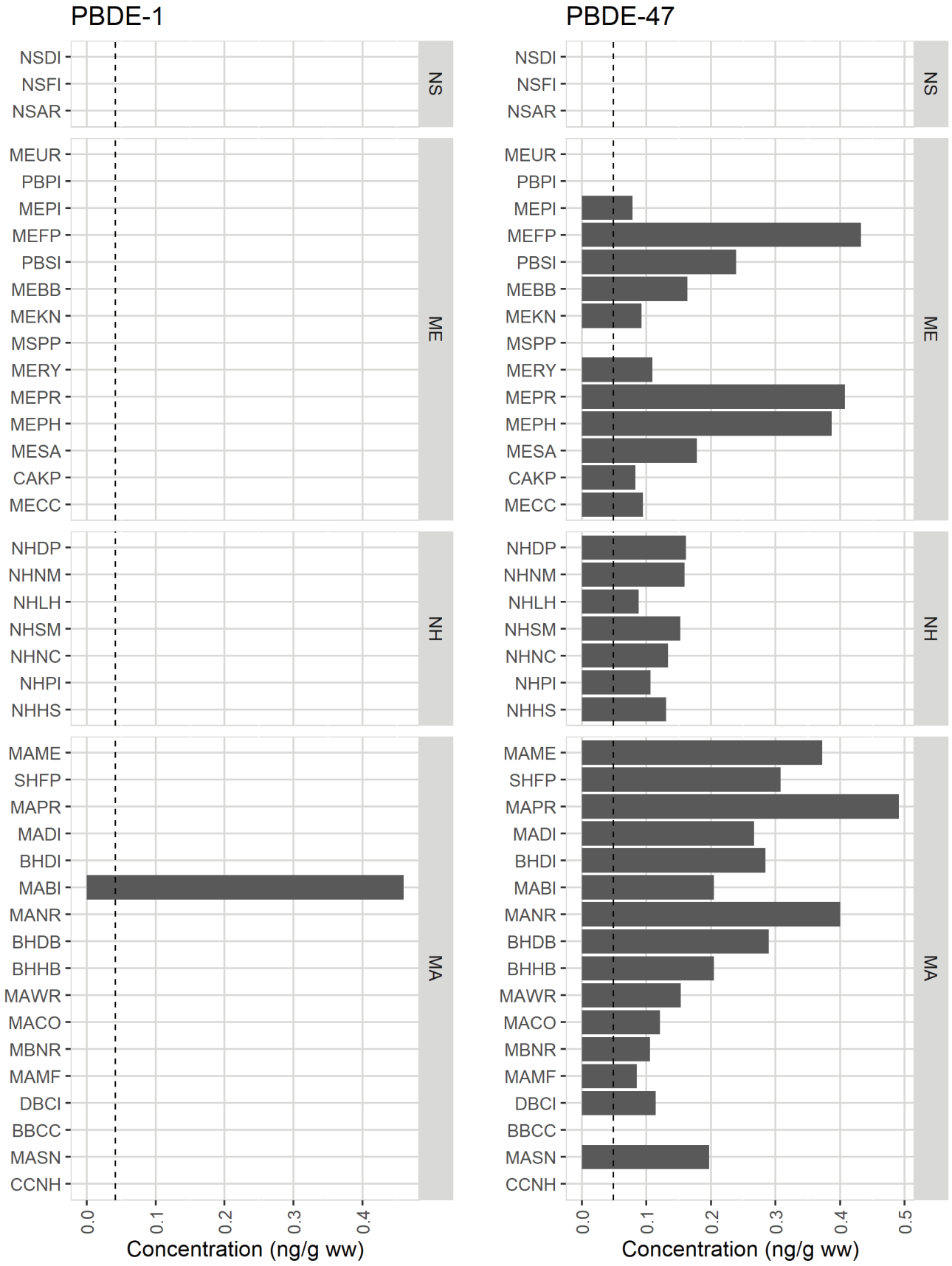

Figure 30. Bar graphs showing magnitude of PBDE contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

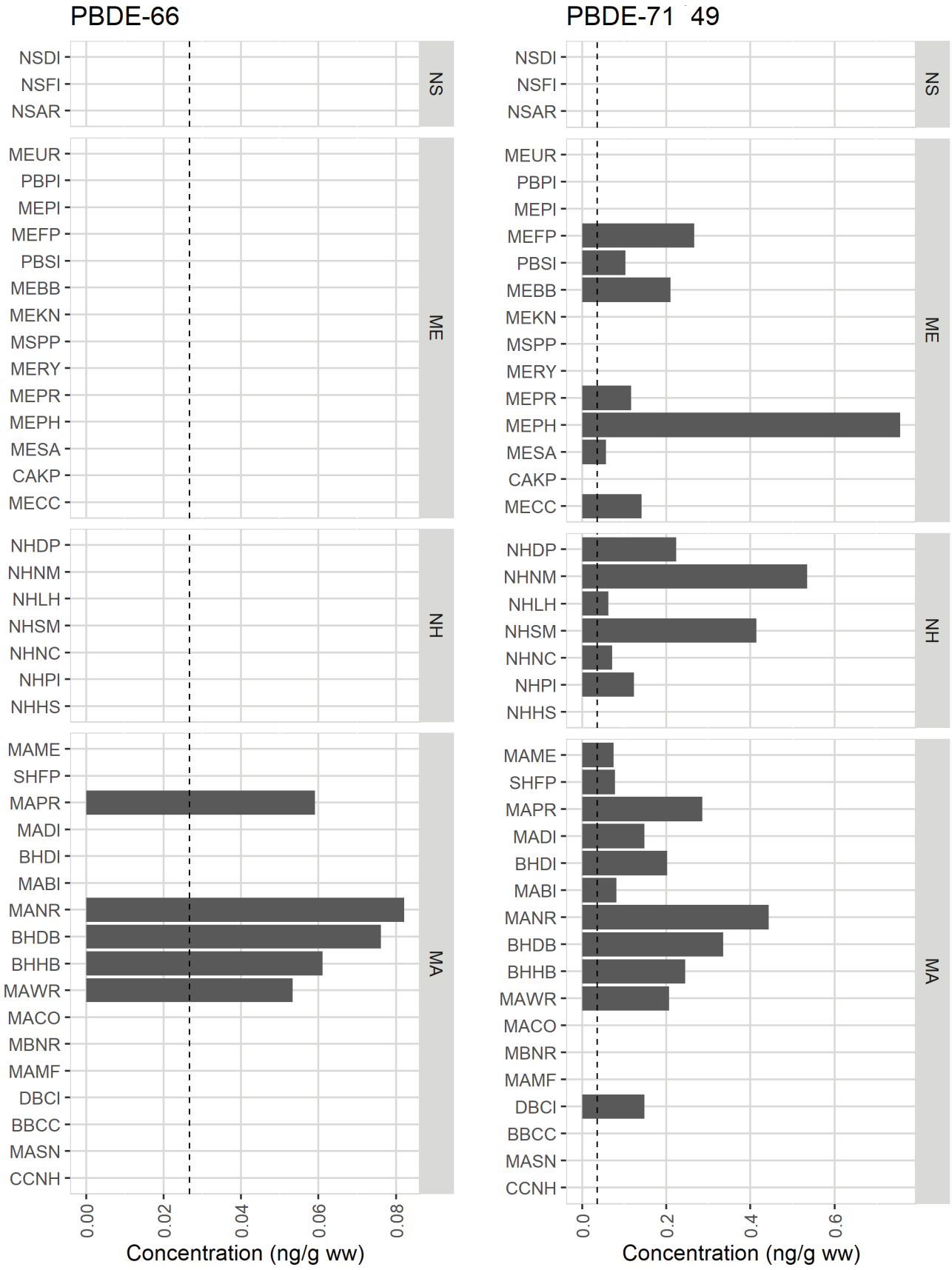

Figure 30. Bar graphs showing magnitude of PBDE contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

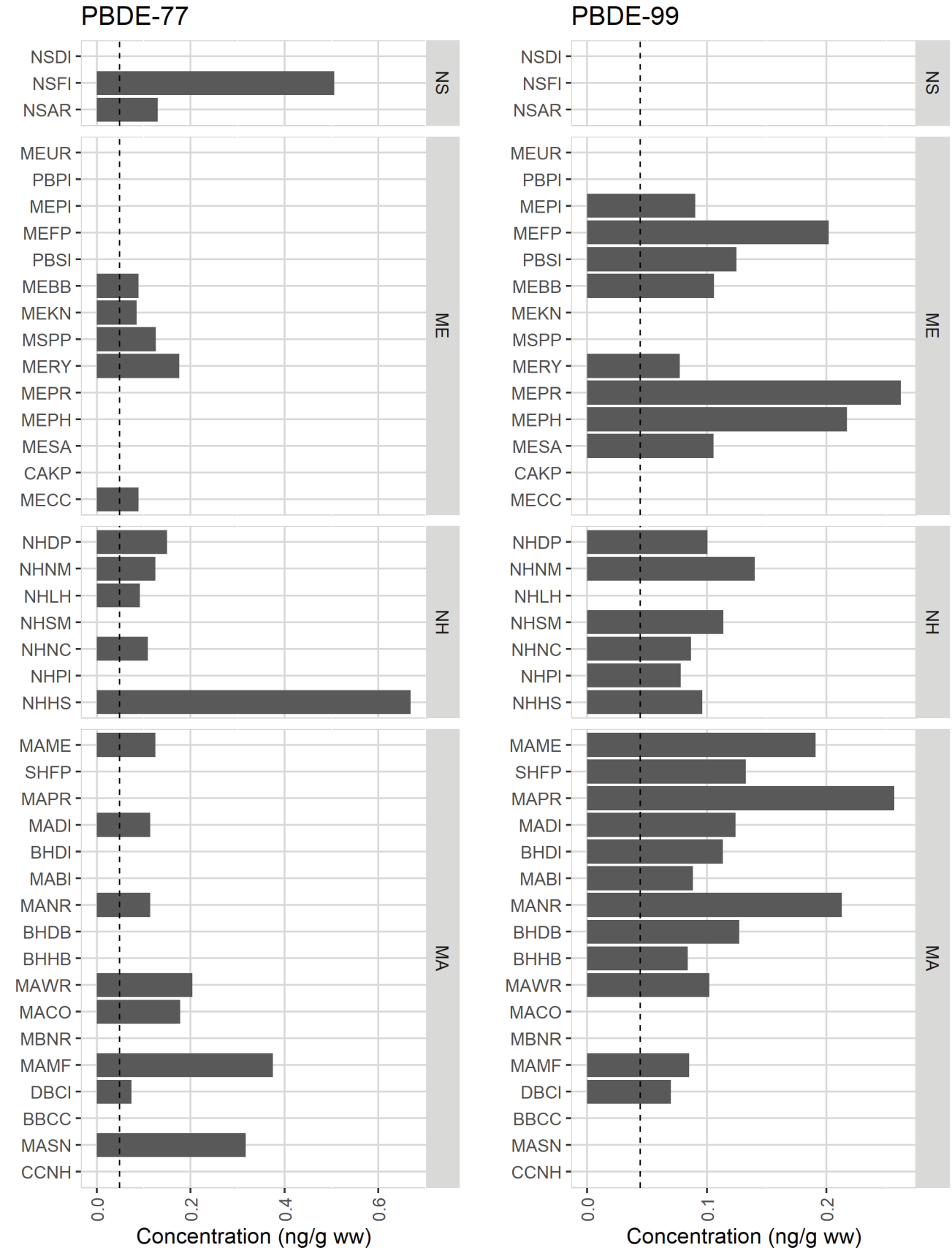

Figure 30. Bar graphs showing magnitude of PBDE contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

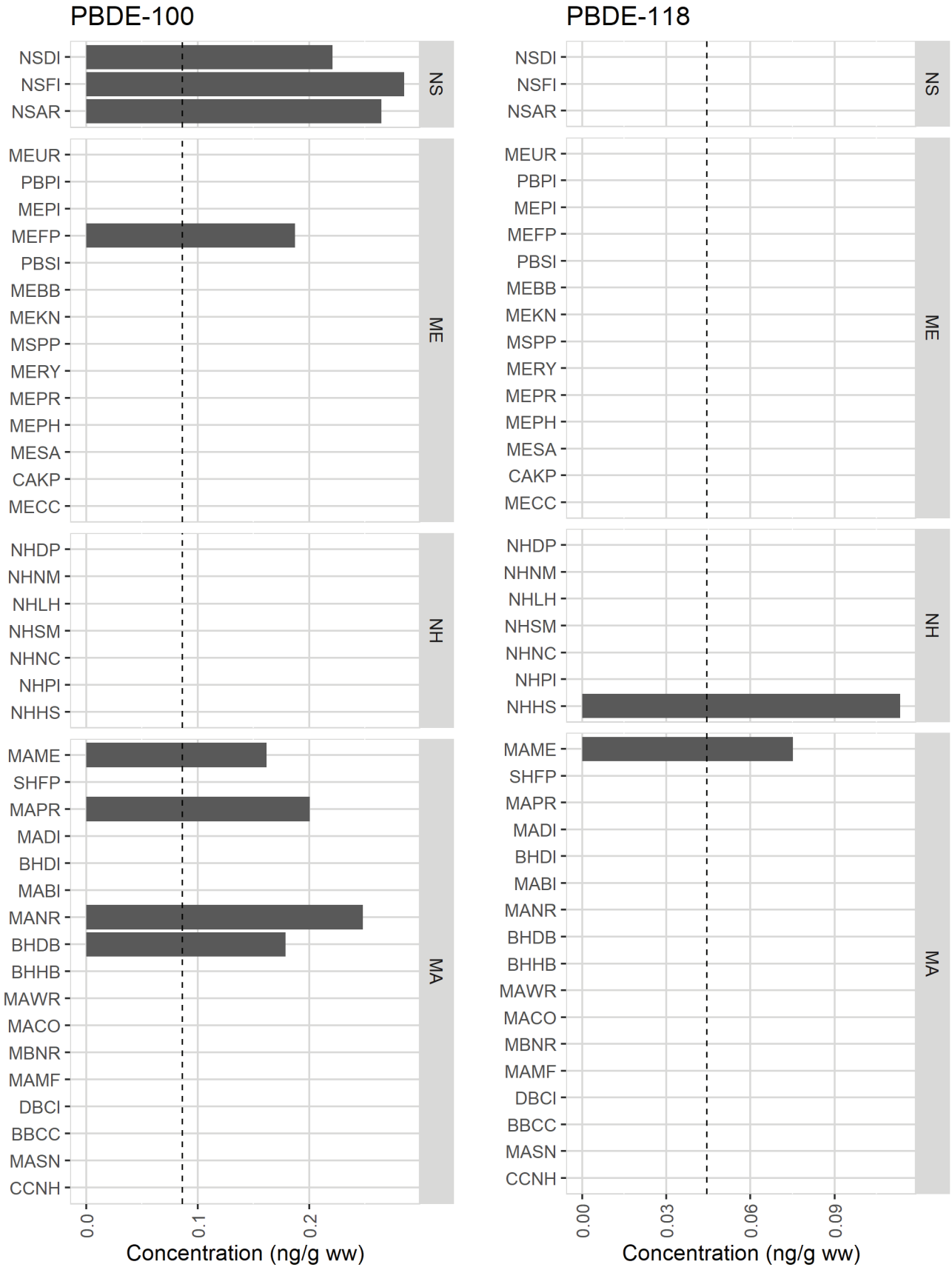

Figure 30. Bar graphs showing magnitude of PBDE contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

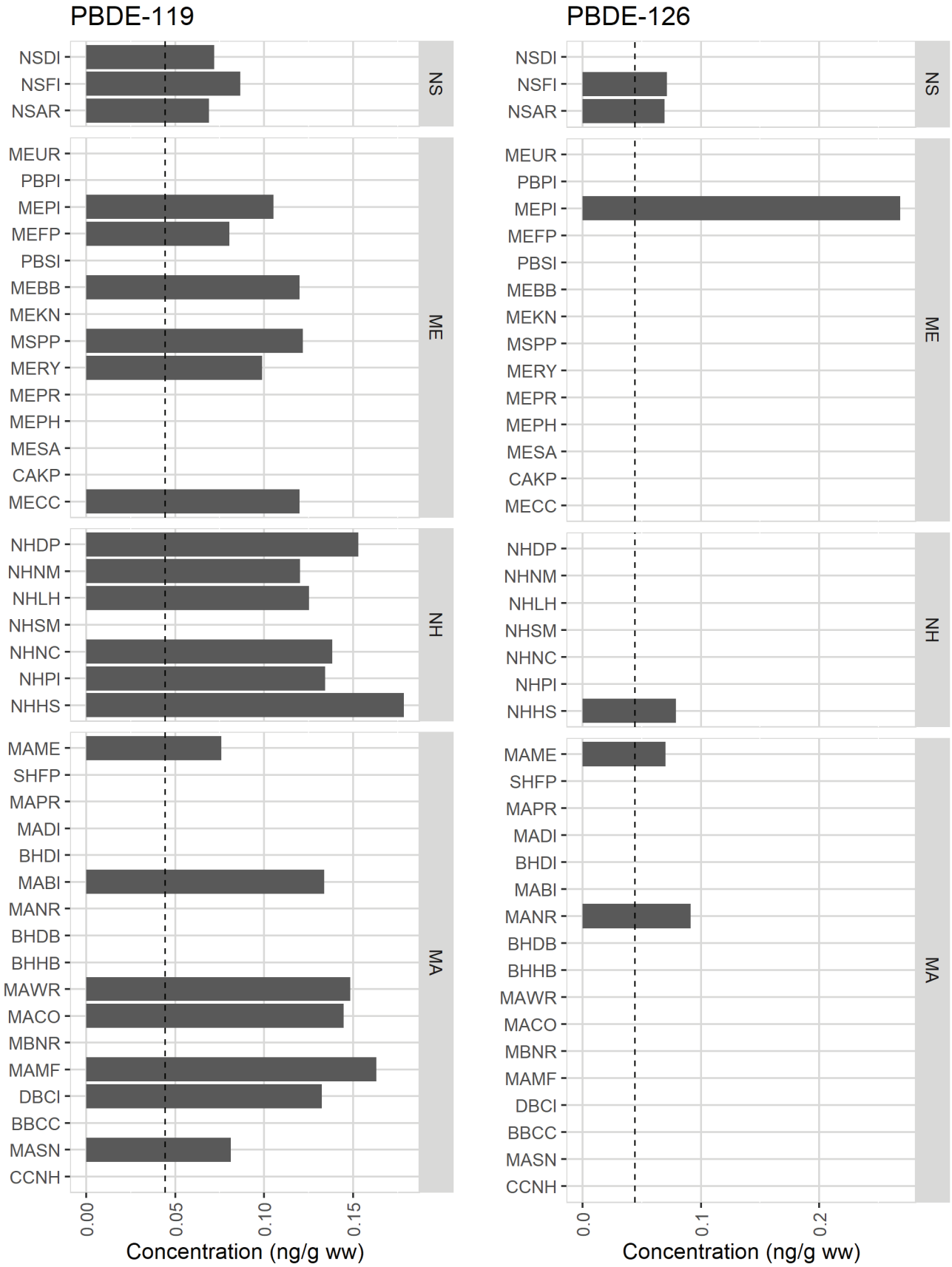

Figure 30. Bar graphs showing magnitude of PBDE contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

GULF-WIDE ASSESSMENT

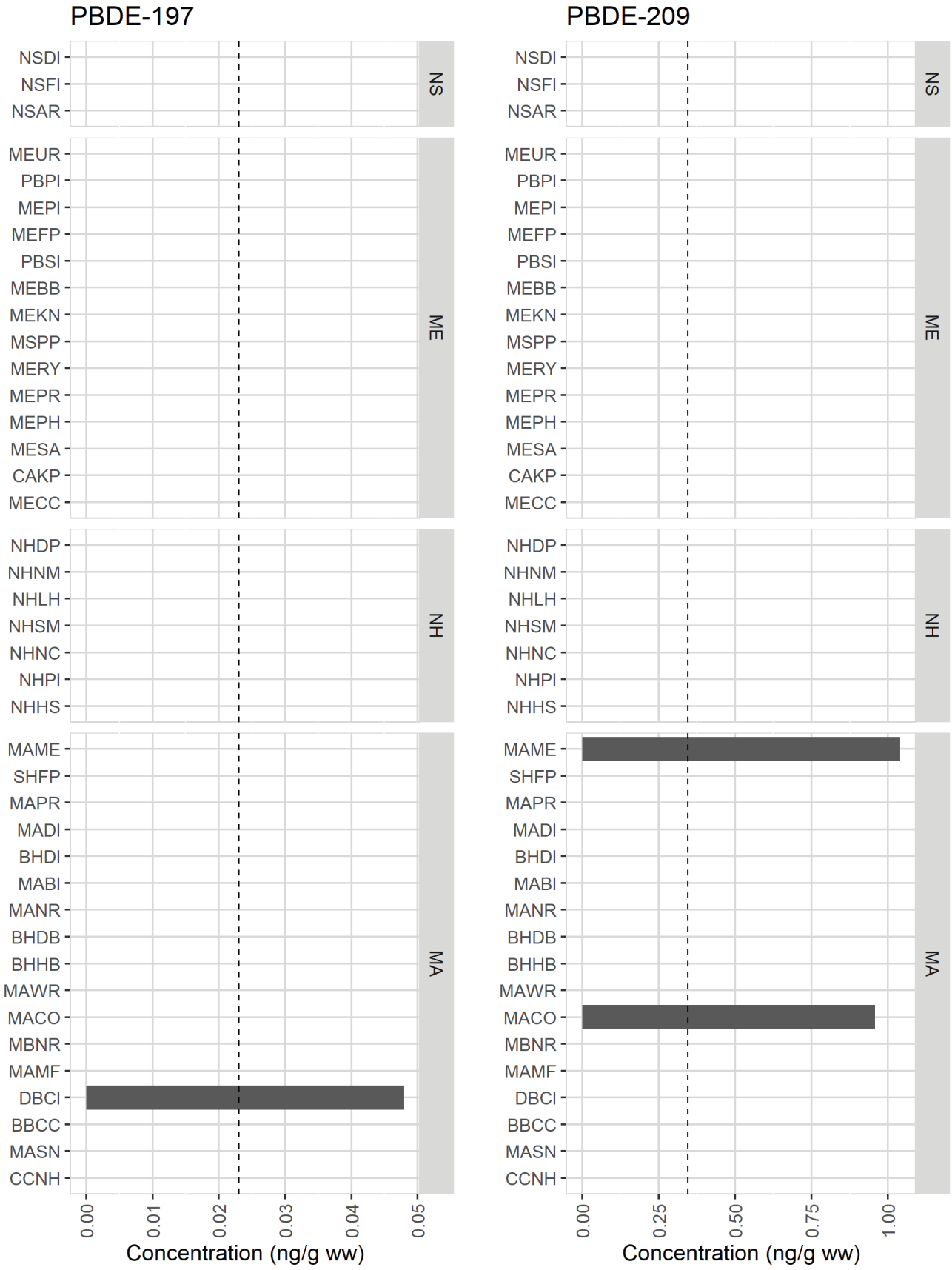

Figure 30. Bar graphs showing magnitude of PBDE contaminants detected in the Gulf of Maine. Dotted line represents the minimum weight corrected detection limit. Sites are listed geographically from north to south, following the coastline.

Summary of PBDEs in mussel tissue

- Polybrominated diphenyl ethers (PBDEs) were analyzed in a total of 41 mussel tissue samples (17 in MA, 14 in ME, 7 in NH and 3 in NS).
- Twelve of the 51 PBDE contaminants were detected Gulf-wide (Figure 29).
- The most frequently detected PBDEs in the Gulf of Maine were the PBDE-47 congener with a Gulf-wide detection frequency of 80.5%, followed by PBDE-99 (63.4%), PBDE-71/49 (58.5%), PBDE-119 (53.66%) and PBDE-77 (48.8%) (Table 31).
- The PBDE-47 congener was found at a total of 33 monitoring sites including sites in MA, ME, and NH (Figure 29).
- The PBDE-99 congener was found at a total of 26 monitoring sites including sites in MA, ME, and NH.
- The PBDE-71/49 congener was found at a total of 24 monitoring sites including sites in MA, ME, and NH.
- The PBDE-119 congener was found at a total of 22 monitoring sites including sites in MA, ME, NH and NS.
- The PBDE-77 congener was found at a total of 22 monitoring sites including sites in MA, NH, NS, and ME.
- The remainder of the detected PBDE contaminants were found at various numbers at diverse monitoring sites throughout the Gulf of Maine including PBDE-100, PBDE-119, and PBDE-126 congeners found in the NS jurisdiction in Canada (Figures 29 and 30).
- The concentration of PBDE contaminants detected varied greatly in mussel tissue across the Gulf (Figure 30).
- The maximum concentrations found across the study area were recorded for the congener PBDE-209 found at 1.04 ng/g ww and 0.96 ng/g ww at the Merrimack River (MAME) and Cohassett (MACO) sites respectively in MA. The congeners PBDE-71/49 was measured at 0.76 ng/g ww at the Stroudwater-Fore Portland Harbor (MEPH) site in ME, and the congener PBDE-77 was detected at a concentration of 0.67 ng/g ww in mussel sample from the Hampton-Seabrook Estuary (NHHS) site in NH (Figure 30, Appendix 5).
- PBDE-77 was also detected at the concentration of 0.51 ng/g ww at the Minas/Cobequid Shore Five Islands (NSFI) site in NS, Canada (Figure 30, Appendix 5). PBDE-77 was detected at 20 of the 41 monitoring sites in the study area and the PBDE-77 concentration at the NSFI site in Canada was the second highest Gulf-wide (Appendix 5).
- With the exception of four sites, the Cape Cod Nauset Harbor (CCNH) and Buzzards Bay Cape Cod Canal (BBCC) in MA, and Penobscot Bay Pickering Island (PBPI) and Union River (MEUR) in ME, all of the monitoring sites in Gulf of Maine had at least one BFR contaminant detected. Hence, BFR contaminants were ubiquitous in the study area from Cape Cod to the Bay of Fundy in Nova Scotia, Canada. BFR contaminants were detected in all land-use categories including open-water locations such as Penobscot Pickering Island (MEPI) and Merriconeag Sound Potts Point (MSPP) sites in ME (Table 4). Spatially, BFR contaminants were detected more frequently at sites located at the mouth of rivers such as the Merrimack River (MAME) and the Neponset River (MANR) sites and estuaries such as Weir River Estuary (MAWR) and Hampton-Seabrook Estuary (NHHS) (Table 32).

Summary of PBDEs in mussel tissue (cont.)

- Total PBDE detection frequency was not correlated with either land-use category or percent impervious surface. However, some individual PBDE congener concentrations were correlated. PBDE-47, PBDE-71/49, and PBDE-99 concentrations were all positively correlated with percent impervious surface at various buffer sizes but correlation coeffecients were weak or moderate. PBDE-47 concentrations were correlated with impervious surface in the 2,3,4 and 5 km buffers (p=.006 - .029, rho= 0.36 - 0.43). They were also higher at sites in the developed land-use category than the undeveloped category in a 5 km buffer (p=.04). PBDE-71/49 and PBDE-99 were correlated with percent impervious surface in all buffer sizes (p=.001 - <.001, rho= 0.51-0.60; p=.002-.01, rho= 0.41-0.48). PBDE-71/49 concentrations were higher at sites in the developed land-use category than sites in both undeveloped and open-water land-use categories in a 2, 3, 4, and 5 km buffer (p=.034, p=.035, p=.001, p<.001). PBDE-99 concentrations were higher at sites in the developed land-use category than in the undeveloped land-use category in a 3 km buffer as well as higher than sites in both undeveloped and open-water land-use categories in a 5 km buffer (p=.046, p=.002). (Appendix 6).

Jurisdiction-specific assessments: MASSACHUSETTS SUMMARY

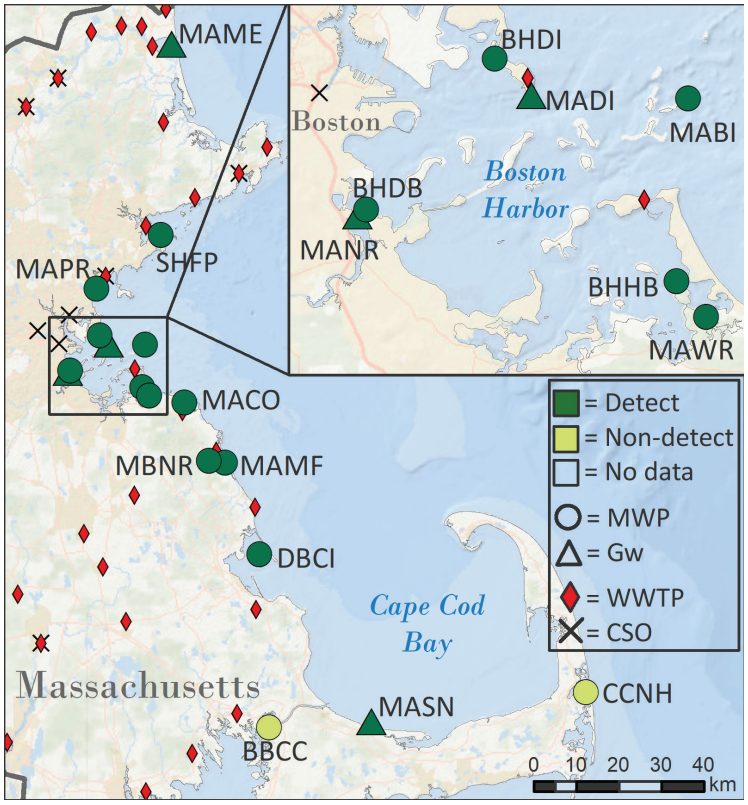

Figure 31. Map of MA jurisdiction highlighting location of sites with PBDE detection in mussel tissue.

Table 33. PBDE compounds frequency of detection in mussel tissue from MA jurisdiction (n = 17).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PBDE-99	12	17	70.6
PBDE-77	8	17	47.1
PBDE-66	5	17	29.4
PBDE-209	2	17	11.8
PBDE-197	1	17	5.9
PBDE-1	1	17	5.9

Summary of PBDEs in Massachusetts

- Mussel tissue samples from a total of 17 sites were tested for PBDE contaminants in MA (Figure 31).
- PBDE contaminants are widespread in MA with 15 of the 17 monitoring sites having at least one detectable level of PBDE contaminants (Figure 31).
- The most frequently detected PBDE contaminants in MA were the congeners PBDE-47 (88.2%), PBDE-99 (70.6%), PBDE-71/49 (64.7%), and PBDE-77 (47.1%) (Table 33).
- The remainder of the detected PBDE contaminants were found at variable number of sites throughout the MA study area (Table 33).
- The concentrations of PBDE contaminants detected in MA varied greatly. The two highest concentrations among the PBDE contaminants tested were recorded in MA and were both for PBDE-209 with a concentration of 1.04 ng/g ww at the Merrimack River (MAME) monitoring site and 0.96 ng/g ww at the Cohassett (MACO) site (Figure 30, Appendix 5). The next highest concentrations found in MA were for the congeners PBDE-47 (0.49 ng/g ww), PBDE-1 (0.46 ng/g ww), and PBDE-71/49 (0.44) found respectively at the Pines River (MAPR), Boston Harbor Brewster Island (MABI), and Neponset River (MANR) monitoring sites (Appendix 5).
- PBDE contaminants were ubiquitous in coastal water of MA and were detected in developed, undeveloped and open-water land-use categories (Table 4). However, PBDE contaminants in the jurisdiction, were more frequently detected at monitoring sites located around mouths of rivers such as the Merrimack River (MAME) and the Neponset River (MANR) sites, in estuarine locations such as the Weir River Estuary (MAWR) site, and in harbor areas such as the Dorchester Bay (BHDB) and Hingham Bay (BHHB) sites in Boston Harbor (Figure 31). The concentrations of PBDEs in the Boston Harbor area might be linked to the presence of wastewater treatment plants and combined sewer outfalls in the watersheds.

Jurisdiction-specific assessments: NEW HAMPSHIRE SUMMARY

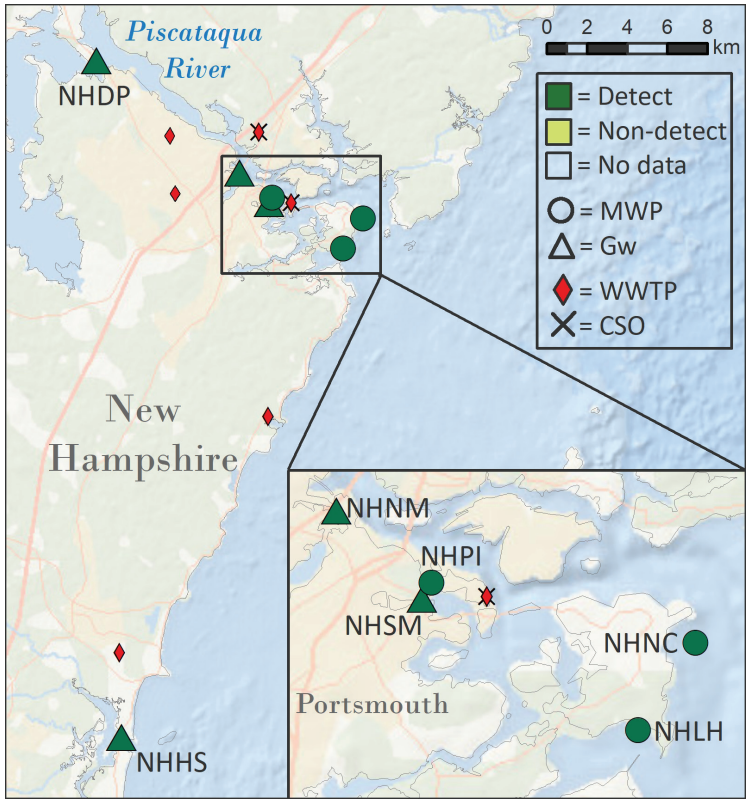

Figure 32. Map of NH jurisdiction highlighting location of sites with PBDE detection in mussel tissue.

Table 34. PBDE compounds frequency of detection in mussel tissue from NH jurisdiction (n = 7).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PBDE-47	7	7	100.0
PBDE-119	6	7	85.7
PBDE-99	6	7	85.7
PBDE-71/49	6	7	85.7
PBDE-77	5	7	71.4
PBDE-126	1	7	14.3
PBDE-118	1	7	14.3

Summary of PBDEs in New Hampshire

- Mussel tissue samples from a total of seven sites were tested for PBDE contaminants in NH (Figure 32).
- PBDE contaminants are wide spread in NH with one or more of the contaminants found at all monitoring sites (Figure 32).
- Seven of the 51 PBDE contaminants tested were found in the NH jurisdiction (Table 34).
- The most frequently detected PBDE contaminants in NH were the congeners PBDE-47 (100%), PBDE-119 (85.7%), PBDE-99 (85.7%), PBDE-71/49 (85.7%) and PBDE-77 (71.4%) (Table 34).
- The remainder of the detected PBDE compounds, PBDE-126 and PBDE-118, were found at one site each in the NH jurisdiction (Table 34).
- The concentrations of PBDE contaminants detected in NH varied from 0.06 for PBDE-71/49 to 0.67 ng/g ww for the PBDE-77 congener at the Piscataqua River Little River (NHLH) and Hampton-Seabrook Estuary (NHHS) monitoring sites respectively (Appendix 5). The next two highest PBDE concentrations in NH were for congener PBDE-71/49 (0.54 ng/g ww and 0.41 ng/g ww) and were found respectively at the North Mill Pond (NHNM) and South Mill Pond (NHSM) monitoring sites (Appendix 5).
- PBDE contaminants were ubiquitous in coastal waters of NH. Based on our land-use classification, PBDEs were detected in developed, undeveloped and open-water land-use categories (Table 4) at each monitoring site in NH (Figure 32). Discharges from wastewater treatment plants and combined sewer outfalls within the coastal watersheds might be the most important source of the presence and wide distribution of PBDE contaminants in the NH coastal monitoring area (Figure 32).

Jurisdiction-specific assessments: MAINE SUMMARY

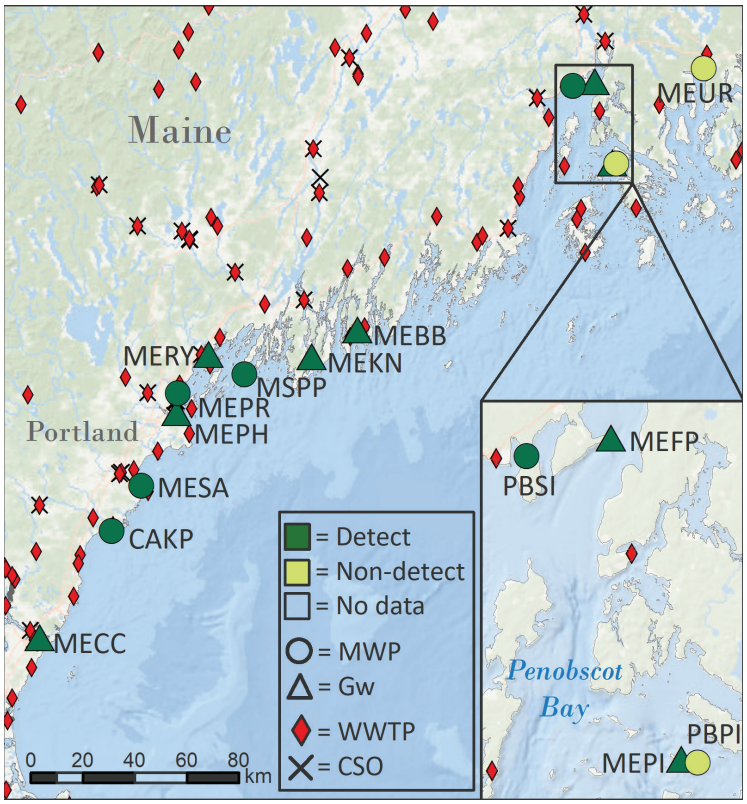

Figure 33. Map of ME jurisdiction highlighting location of sites with PBDE detection in mussel tissue.

Table 35. PBDE compounds frequency of detection in mussel tissue from ME jurisdiction (n = 14).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PBDE-47	11	14	78.6
PBDE-99	8	14	57.1
PBDE-71/49	7	14	50.0
PBDE-119	6	14	42.9
PBDE-77	5	14	35.7
PBDE-126	1	14	7.1
PBDE-100	1	14	7.1

Summary of PBDEs in Maine

- Mussel tissue samples from a total of 14 sites were tested for PBDE contaminants in ME (Figure 33).
- With the exception of the Union River (MEUR) and Penobscot Pickering Island (PBPI) sites, at least one PBDE contaminant was detected at all of the 12 remaining monitoring sites in ME (Figure 33, Appendix 5).
- Seven of the 51 PBDE contaminants tested were found in the ME jurisdiction (Table 35).
- The most frequently detected PBDE contaminants in ME were the congeners PBDE-47 (78.6%), PBDE-99 (57.1%), PBDE-71/49 (50.0%), PBDE-119 (42.9%) and PBDE-77 (35.7%) (Table 35).
- The remainder of the detected PBDE contaminants, PBDE-126 and PBDE-100, were found at one site each throughout the ME study area (Table 35).
- The concentration of PBDE contaminants detected in ME varied from 0.06 to 0.76 ng/g ww, both for the PBDE-71/49 congener at the Saco River (MESA) and Stroudwater-Fore Portland Harbor (MEPH) monitoring sites respectively (Figure 30, Appendix 5).
- PBDE contaminants were present at 12 of the 14 monitoring sites in ME and were detected in developed, undeveloped and open-water land-use categories (Table 4). Discharges from wastewater treatment plants and combined sewer outfalls within the coastal watersheds might be the most important source of PBDE contaminants in the ME coastal monitoring area (Figure 33).

Jurisdiction-specific assessments: Nova Scotia Summary

Figure 34. Map of NS jurisdiction highlighting location of sites with PBDE detection in mussel tissue.

Table 36. PBDE compounds frequency of detection in mussel tissue from NS jurisdiction (n = 3).

Compound	Number of Detects	Number of Sample Sites	Frequency (%)
PBDE-119	3	3	100.0
PBDE-100	3	3	100.0
PBDE-126	2	3	66.7
PBDE-77	2	3	66.7

Summary of PBDEs in Nova Scotia

- Mussel tissue samples from a total of three sites were tested for PBDE contaminants in NS (Figure 34).
- At least one PBDE contaminant was detected at each of the three monitoring sites in NS (Figure 34).
- Four of the 51 PBDE contaminants were detected in the NS jurisdiction (Table 36).
- PBDE-119 and PBDE-100 were detected at the Chignecto Bay Apple River (NSAR), Annapolis Basin Digby (NSDI), and Minas/Cobequid Shore Five Islands (NSFI) sites. Both congeners PBDE-77 and PBDE-126 were detected at the NSFI and NSAR monitoring sites (Figure 30, Appendix 5).
- The concentration of 0.51 ng/g ww for PBDE-77 at the NSFI monitoring site was the highest concentration found among the PBDE contaminants detected in NS (Appendix 5). The lowest concentration of 0.07 ng/g ww was found for PBDE-119 and PBDE-126 at NSAR, and NSFI and NSAR respectively (Appendix 5). The congener PBDE-100 had similar concentrations between 0.22 ng/g ww and 29 ng/g ww at all three NS monitoring sites (Appendix 5).
- Based on our land-use classification, there was no land-use type associated to the NS monitoring sites (Table 4). However, the detection of PBDE contaminants in the coastal water of Nova Scotia is another testament of the environmental ubiquity of BFR contamination.

Summary

Gulf of Maine sunset. Credit: NOAA

Summary

In collaboration with the Gulf of Maine Gulfwatch (Gw) Program, the national Mussel Watch Program (MWP) conducted an assessment of the magnitude and distribution of contaminants of emerging concern (CECs) in coastal waters of the Gulf of Maine. Using blue mussels (*Mytilus* species) as indicators of water contamination, samples were assessed for alkylphenol compounds (APs), alternative flame retardants (AFRs), current-use pesticides (CUPs), per- and polyfluoroalkyl substances (PFASs), pharmaceutical and personal care products (PPCPs), and polybrominated flame retardants (BFRs) such as polybrominated biphenyls (PBBs) and polybrominated diphenyl ethers (PBDEs). The mussel samples were collected at a combination of historic MWP and Gw monitoring sites located across four jurisdictions of the Gulf of Maine: Maine, Massachusetts, New Hampshire, and Nova Scotia (Canada). Site selection for this study was conducted in collaboration with resource managers in the region that are part of the Gulf of Maine Council on Marine Environment and it involved a strategic mixture of sites that met both programs' monitoring needs. Sample collection was conducted by the Gw Program following modified standard protocols utilized by the national MWP and the Gw program (Apeti et al., 2012). Mussel samples from a combined 41 monitoring sites were measured for a total of 249 individual CEC compounds, including 4 APs, 9 AFRs, 33 CUPs, 12 PFASs, 121 PPCPs, and 70 BFRs. The following provide a succinct summary of the findings of each class of CEC.

The magnitude of AP contaminants, used in detergents and surfactants in industrial processes, varied in mussel tissue across the Gulf of Maine (Figure 5). The AP contaminants 4-nonylphenol mono-ethoxylate (NP1EO), 4-nonylphenol di-ethoxylate (NP2EO), and 4-n-octylphenol (4-n-OP) were detected. NP1EO was the most frequently detected compound with a maximum concentration of 16.5 ng/g ww recorded at the South Mill Pond (NHSM) site in NH (Figure 7, Appendix 1). Jurisdiction specific assessments indicated that APs were more prevalent in NH and ME than MA. AP contaminants were not detected in NS, Canada. Published data by Apeti et al. (2018) and Kimbrough et al. (2018) have also reported the bioaccumulation of AP contaminants concentration above detection limits respectively in oyster tissues in the Chesapeake Bay and dreissenid mussel tissues in the Great Lakes.

Site MBNR. Credit: NOAA

Summary

Site MBNR. Credit: NOAA

AFR contaminants, which are primarily used in household consumer products such as upholstery, polystyrene and textiles, were only detected in the mussel tissue from MA and ME. Among the AFRs measured, only the TBB and TBPH contaminants were detected. Similar observations were reported in an AFR assessment in oyster tissue from the Chesapeake Bay (Apeti et al., 2018). A maximum concentration of 3.27 ng/g ww was recorded in the mussel sample from the MADI site in MA for the TBB contaminant, while the TBPH contaminant was found at a concentration of 0.73 ng/g ww mussels from MEKN site in ME (Figure 12, Appendix 2).

CUPs include pesticides and their associated degradation products. These pesticides are typically designed to be more water-soluble than the legacy organochlorine pesticides and often do not readily bioaccumulate in organisms. The results indicated that CUP contaminants were not detected in the Gulf of Maine. However, these contaminants have been measured in oysters (Apeti et al. 2018) and freshwater invasive dreissenid mussels (Kimbrough et al., 2018) at low concentrations relative to detection limit values. This indicates that CUPs can potentially bioaccumulate in coastal organisms, but accumulation magnitude may depend on location and land-use types.

Per- and polyfluoroalkyl substances (PFASs) are industrial chemicals related to surface protection/coatings and fire fighting foam. Among the PFASs, contaminants were detected at different locations across the Gulf of Maine including the toxic perfluorooctane sulfonate (PFOS) (Li, 2008). However, similar to a previously published study by Apeti et al. (2018) on oysters, the most pervasive PFAS in blue mussels was perfluorooctane sulfonamide (PFOSA), which was found in all Gulf of Maine jurisdictions except NS in Canada. A maximum concentration of 5.46 ng/g ww was recorded for PFOSA at the MEPH in Maine, and perfluorooctane sulfonate (PFOS) found at the MANR site in MA had a concentration of 0.60 ng/g ww (Figure 18, Appendix 3).

Table 37. Summary of Gulf-wide number of detects measured at each site ranked by Juridiction and Total detetection frequency (%).

Jurisdiction	Site	Total number of compounds analyzed	Total number of compounds detected	Total detection frequency (%)	AP Total	AFR Total	CUP Total	PFAS Total	PPCP Total	PBB Total	PBDE Total
MA	MANR	249	17	6.8	1	0	0	2	6	0	7
MA	MAME	249	15	6.0	1	0	0	1	4	0	9
MA	BHDB	248	10	4.0	0	0	0	0	5	0	5
MA	BHDI	249	10	4.0	1	1	0	0	5	0	3
MA	MAPR	249	9	3.6	0	0	0	0	4	0	5
MA	MADI	249	9	3.6	0	1	0	1	3	0	4
MA	MABI	239	8	3.3	0	NA	0	1	2	0	5
MA	DBCI	248	8	3.2	0	1	0	0	1	0	6
MA	MAWR	248	8	3.2	0	0	0	1	1	0	6
MA	SHFP	248	7	2.8	0	0	0	0	4	0	3
MA	BHHB	249	7	2.8	0	1	0	0	2	0	4
MA	MACO	248	6	2.4	0	0	0	1	1	0	4
MA	MAMF	248	5	2.0	0	0	0	0	1	0	4
MA	MASN	249	4	1.6	0	0	0	0	1	0	3
MA	BBCC	249	3	1.2	0	0	0	1	2	0	0
MA	MBNR	249	3	1.2	0	0	0	0	2	0	1
MA	CCNH	248	2	0.8	0	0	0	1	1	0	0
MA Total		**4216**	**130**	**3.1**	**3**	**4**	**0**	**9**	**45**	**0**	**69**
ME	MECC	249	11	4.4	1	0	0	1	5	0	4
ME	MEPR	249	10	4.0	2	1	0	0	4	0	3
ME	MEKN	249	10	4.0	1	2	0	1	4	0	2
ME	MEBB	249	10	4.0	0	0	0	2	3	0	5
ME	MEPH	249	8	3.2	1	0	0	1	3	0	3
ME	MESA	249	7	2.8	0	0	0	0	4	0	3
ME	MERY	249	7	2.8	0	0	0	0	3	0	4
ME	MEFP	249	7	2.8	0	0	0	0	2	0	5
ME	MSPP	248	5	2.0	0	0	0	0	3	0	2
ME	PBSI	249	5	2.0	0	0	0	0	2	0	3
ME	MEPI	249	5	2.0	0	0	0	0	1	0	4
ME	CAKP	249	3	1.2	1	0	0	0	1	0	1
ME	PBPI	249	3	1.2	0	0	0	0	3	0	0
ME	MEUR	249	2	0.8	0	0	0	0	2	0	0
ME Total		**3485**	**93**	**2.7**	**6**	**3**	**0**	**5**	**40**	**0**	**39**
NH	NHHS	249	13	5.2	2	0	0	0	5	0	6
NH	NHNM	240	11	4.6	1	NA	0	1	4	0	5
NH	NHSM	249	11	4.4	2	0	0	1	5	0	3
NH	NHDP	249	11	4.4	1	0	0	0	5	0	5
NH	NHNC	248	10	4.0	1	0	0	1	3	0	5
NH	NHPI	248	8	3.2	0	0	0	0	4	0	4
NH	NHLH	248	6	2.4	0	0	0	1	1	0	4
NH Total		**1731**	**70**	**4**	**7**	**0**	**0**	**4**	**27**	**0**	**32**
NS	NSFI	70	4	5.7	NA	NA	NA	NA	NA	0	4
NS	NSAR	248	4	1.6	0	0	0	0	0	0	4
NS	NSDI	248	3	1.2	0	0	0	0	1	0	2
NS Total		**566**	**11**	**1.9**	**0**	**0**	**0**	**0**	**1**	**0**	**10**

Summary

Environmental PPCPs include a wide spectrum of therapeutic and consumer-use compounds such as prescription and over-the-counter medications, hormones, synthetic fragrances, disinfectants, insect repellants, and antimicrobial agents. PPCP contaminants most frequently detected were the insect repellant DEET, the antidepressant drug sertraline, and the antihistamine drug diphenhydramine. A similar pattern was observed by Kimbrough et al. (2018) with DEET, sertraline, and diphenhydramine being among the most commonly detected PPCPs in the Great lakes. It is worth noting that some PPCPs were found at relatively higher concentrations compared to the others. These include meprobamate, a sedative drug used for insomnia and psychiatric anxiety, and caffeine found at the concentrations of 59.44 and 57.72 ng/g ww respectively at the MABI and SHFP sites in MA. Metprolol and propranolol, which are both used to treat angina and hypertension, were detected respectively at 46.65 and 42.57 ng/g ww in mussel tissues from the sites MERY in ME and NHDP in NH (Figure 24, Appendix 4). PPCP contaminants were indiscriminately found in every jurisdiction in the Gulf of Maine including NS in Canada, however, they were found at higher frequencies and concentrations in harbor areas and near wastewater treatment plants and outfalls.

BFRs, such as PBDEs and PBBs, are toxic firefighting materials with 209 possible unique congeners each. In this study a combined 70 congeners were measured. In contrast to the PBB congeners, which were not detected in any mussel sample, several PBDE congeners were found at various concentrations throughout the Gulf of Maine including all three of the NS sites in Canada. The results showed that the most frequently detected PBDEs in the Gulf of Maine blue mussels were congeners PBDE-47 (at 80.5% of the sites), PBDE-99 (63.4%), PBDE-71/49 (58.5%), PBDE-119 (53.66%) and PBDE-77 (48.8%). These findings mirrored the result of the BFR assessment in oyster tissue from the Chesapeake Bay with virtually the same PBDE congener signature for the most frequently detected PBDEs (Apeti et al., 2018). The maximum concentrations found across the study area were recorded for the congener PBDE-209 found at 1.04 ng/g ww and 0.96 ng/g ww at the Merrimack River (MAME) and Cohassett (MACO) sites respectively in MA. The congeners PBDE-71/49 was measured at 0.76 ng/g ww at the Stroudwater-Fore Portland Harbor (MEPH) site in ME, and the congener PBDE-77 was detected at a concentration of 0.67 ng/g ww in mussel sample from the Hampton-Seabrook Estuary (NHHS) site in NH (Figure 30, Appendix 5).

Table 38. Summary of Gulf-wide detection frequency for each class of CEC assessed in the Gulf of Maine.

Compound Class	Number of Detects	Number of Possible Detects	Detection Frequency (%)
AP	16	160	10
PBDE	150	2091	7.2
PFAS	18	480	3.8
PPCP	113	4838	2.3
AFR	7	342	2
CUP	0	1308	0
PBB	0	779	0

The results indicated that CECs are present at various degrees in coastal waters of the Gulf of Maine and they are being accumulated at various concentrations in coastal resources. Mussel samples from all 41 monitoring sites exhibited the presence of at least two CEC compounds highlighting the ubiquity of these contaminants in the coastal zone throughout the four Gulf of Maine jurisdictions (Table 37). APs had the highest detection frequency at 10%, followed by PBDEs (7.2%) and PFAS (3.8%) (Table 38). It is important to note that the presence and magnitude, hence bioaccumulation of the CEC contaminants in organisms such as mussels are typically compound dependent, with a small subset of contaminants representing the majority of detections within each class.

The distribution and magnitude of the CEC contaminants also depended on location and land-use types in watersheds adjacent to the monitoring location. Based on our land-use assessment, CEC contami-

nants were detected at sites with land-uses categorized as developed, undeveloped and open-water. However, many of the highest detection frequencies were located in developed areas including Boston, MA, Portsmouth, NH, and Portland, ME. Developed land-use and high percent impervious were positively correlated with AP, PFAS and PPCP detection frequencies, as well as the concentrations of several individual compounds (NP1EO, PFOSA, diphenhydramine, sertraline, PBDE-47, PBDE-71/49, and PBDE-99). Total CEC detection frequencies were positively correlated with percent impervious surface in every buffer size but the coefficients of determination were weak (Appendix 6). Total CEC detection frequencies at sites in the developed land-use category were higher than sites in the undeveloped category in both the 2 and 5 km buffer size ($p=0.045$, $p=0.025$).

Many of the contaminant concentrations and detection frequencies showed no discernible correlation with the land-use parameters analyzed in this report. Additionally, when a correlation was determined, correlation coefficients were weak, emphasizing the complexity and variability of the data. Both local influences and those higher up the watershed or stream may be affecting the presence and concentration of contaminants at sites closer to the coastline. For example, higher detection frequencies were located not only in developed areas but also at sites at the mouths of major rivers, like the Merrimack and Kennebec rivers, the latter including drainage from both the Androscoggin and Kennebec rivers. Additionally, a visual assessment suggests that sites with higher detection frequencies and concentrations of CECs were influenced either by wastewater treatment plants or by combined sewer outfalls.

The influence of both anthropogenic and environmental factors makes it difficult to accurately predict the presence and concentration of CEC compounds in the environment. However, as this study shows, they are present and bioccumulating to various degrees in coastal bivalves. This study provides needed data and information for the national MWP and supports water quality data required by coastal resources managers as they develop effective long-term policies protecting services provided by the coastal environment within this region.

Site CAKP. Credit: NOAA

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Appendix 1. AP compound concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	4-n-OP	NP1EO	NP2EO
Minimum MDL		0.41	4.08	4.08
NSDI	NS	0	0	0
NSAR	NS	0	0	0
MEUR	ME	0	0	0
PBPI	ME	0	0	0
MEPI	ME	0	0	0
MEFP	ME	0	0	0
PBSI	ME	0	0	0
MEBB	ME	0	0	0
MEKN	ME	0	7.75	0
MSPP	ME	0	0	0
MERY	ME	0	0	0
MEPR	ME	0	14.9	5.35
MEPH	ME	0	8.97	0
MESA	ME	0	0	0
CAKP	ME	0	5.81	0
MECC	ME	0	6.82	0
NHDP	NH	0	11.3	0
NHNM	NH	0	11.7	0
NHLH	NH	0	0	0
NHSM	NH	0	16.5	6.89
NHNC	NH	0	8.89	0
NHPI	NH	0	0	0
NHHS	NH	1.44	8.80	0
MAME	MA	0	5.99	0
SHFP	MA	0	0	0
MAPR	MA	0	0	0
MADI	MA	0	0	0
BHDI	MA	0	5.55	0
MABI	MA	0	0	0
MANR	MA	0	6.25	0
BHDB	MA	0	0	0
BHHB	MA	0	0	0
MAWR	MA	0	0	0
MACO	MA	0	0	0
MBNR	MA	0	0	0
MAMF	MA	0	0	0
DBCI	MA	0	0	0
BBCC	MA	0	0	0
MASN	MA	0	0	0
CCNH	MA	0	0	0

Appendix 2. AFR compound concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	TBB	TBPH
Minimum MDL		0.17	0.17
NSDI	NS	0	0
NSAR	NS	0	0
MEUR	ME	0	0
PBPI	ME	0	0
MEPI	ME	0	0
MEFP	ME	0	0
PBSI	ME	0	0
MEBB	ME	0	0
MEKN	ME	1.91	0.73
MSPP	ME	0	0
MERY	ME	0	0
MEPR	ME	0.55	0
MEPH	ME	0	0
MESA	ME	0	0
CAKP	ME	0	0
MECC	ME	0	0
NHDP	NH	0	0
NHLH	NH	0	0
NHSM	NH	0	0
NHNC	NH	0	0
NHPI	NH	0	0
NHHS	NH	0	0
MAME	MA	0	0
SHFP	MA	0	0
MAPR	MA	0	0
MADI	MA	3.27	0
BHDI	MA	0.59	0
MANR	MA	0	0
BHDB	MA	0	0
BHHB	MA	1.27	0
MAWR	MA	0	0
MACO	MA	0	0
MBNR	MA	0	0
MAMF	MA	0	0
DBCI	MA	1.56	0
BBCC	MA	0	0
MASN	MA	0	0
CCNH	MA	0	0
MASN	MA	0	0
CCNH	MA	0	0

Appendix 3. PFAS concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	PFOA	PFOS	PFOSA
Minimum MDL		0.12	0.16	0.12
NSDI	NS	0	0	0
NSAR	NS	0	0	0
MEUR	ME	0	0	0
PBPI	ME	0	0	0
MEPI	ME	0	0	0
MEFP	ME	0	0	0
PBSI	ME	0	0	0
MEBB	ME	0.35	0	1.39
MEKN	ME	0	0	2.51
MSPP	ME	0	0	0
MERY	ME	0	0	0
MEPR	ME	0	0	0
MEPH	ME	0	0	5.46
MESA	ME	0	0	0
CAKP	ME	0	0	0
MECC	ME	0	0	0.95
NHDP	NH	0	0	0
NHNM	NH	0	0	2.79
NHLH	NH	0	0	0.71
NHSM	NH	0	0	1.17
NHNC	NH	0	0	1.18
NHPI	NH	0	0	0
NHHS	NH	0	0	0
MAME	MA	0	0	1.21
SHFP	MA	0	0	0
MAPR	MA	0	0	0
MADI	MA	0	0	0.71
BHDI	MA	0	0	0
MABI	MA	0	0	0.29
MANR	MA	0	0.60	0.83
BHDB	MA	0	0	0
BHHB	MA	0	0	0
MAWR	MA	0	0	1.61
MACO	MA	0	0	1.12
MBNR	MA	0	0	0
MAMF	MA	0	0	0
DBCI	MA	0	0	0
BBCC	MA	0	0	0.42
MASN	MA	0	0	0
CCNH	MA	0	0	0.24

Appendix 4. PPCP compound concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	17β-estradiol	Amitriptyline	Androstenedione	Atenolol	Azithromycin	Benzoylecgonine	Benztropine	Caffeine	Carbadox	Carbamazepine	Cimetidine
Minimum MDL		0.49	0.05	0.49	0.07	0.12	0.05	0.05	1.22	0.12	0.12	0.07
NSDI	NS	0	0	0	0	0	0	0	0	0	0	0
NSAR	NS	0	0	0	0	0	0	0	0	0	0	0
MEUR	ME	0.74	0	0	0	0	0	0	0	0	0	0
PBPI	ME	0	0	0	0	0	0	0	0	0	0	0
MEPI	ME	0	0	0	0	0	0	0	0	0	0	0
MEFP	ME	0	0	0	0	1.16	0	0	0	0	0	0
PBSI	ME	0	0	0	0	0	0	0	0	0	0	0
MEBB	ME	0	0	0	0	0	0	0	0	0	0	0
MEKN	ME	0	0	0	0	0	0	0	0	0	0	0
MSPP	ME	0	0	0	0	0	0	0	0	0	0	0
MERY	ME	0	0	0	0	0	0	0	0	0	0	0
MEPR	ME	0	0	0	0	0	0	0	0	0	0	0
MEPH	ME	0	0	0	0	0	0	0.16	0	0	0	0
MESA	ME	0.56	0	0	0	0	3.61	0	0	0	0	0
CAKP	ME	0	0	0	0	0	0	0	0	0	0	0
MECC	ME	0	0	0	0	0	0	0	0	0	0	5.75
NHDP	NH	0	0	0	0	0	0	0	0	0	0	0
NHNM	NH	0	0	0	0	0	0	0	0	0	0	0
NHLH	NH	0	0	0	0	0	0	0	0	0	0	0
NHSM	NH	0	2.35	0	0	0	0	0	0	0	0	0
NHNC	NH	0	0	0	0	0	0	0	0	0	0	0
NHPI	NH	0	0	0	0	0	0	0	0	0	0	0
NHHS	NH	0	0	0	1.82	0	0	0	0	0	0	0
MAME	MA	0	0	0	0	0	0	0	0	0	0	0
SHFP	MA	0	0	0	0	0	0	0	57.7	0	0	0
MAPR	MA	0	0	0	0	0	0	0	0	0	0	0
MADI	MA	0	0	0	0	0	0	0	0	0	0	0
BHDI	MA	0	0	0	0	0	0	0	0	0	0	0
MABI	MA	0	0	0	0	0	0	0	0	0	0	0
MANR	MA	0	0	0	0	0	0	0	0	0	1.76	0
BHDB	MA	0	0	0	0	0	0	0	0	145	0	0
BHHB	MA	0	0	0	0	0	0	0	0	0	0	0
MAWR	MA	0	0	0	0	0	0	0	0	0	0	0
MACO	MA	0	0	0	0	0	0	0	0	0	0	0
MBNR	MA	0	0	0	0	0	0	0	0	0	0	0
MAMF	MA	0	0	0	0	0	0	0	0	0	0	0
DBCI	MA	0	0	0	0	0	0	0	0	0	0	0
BBCC	MA	0	0	0	0	0	0	0	0	0	0	0
MASN	MA	0	0	14.6	0	0	0	0	0	0	0	0
CCNH	MA	0	0	0	0	0	0	0	0	0	0	0

Appendix 4 (cont.). PPCP compound concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	Citalopram	Cocaine	Cotinine	DEET	Diazepam	Diphenhydramine	Fluoxetine	Hydrocortisone	Meprobamate	Mestranol	Metprolol
Minimum MDL		0.15	0.02	0.17	0.12	0.05	0.07	0.12	0.98	0.24	1.95	0.24
NSDI	NS	0	0	0	0	0	0	0	0	0	0	0
NSAR	NS	0	0	0	0	0	0	0	0	0	0	0
MEUR	ME	0	0	0	0	0	0	0	0	0	0	0
PBPI	ME	0	0	0	17.5	0	0	0	0	0	0	0
MEPI	ME	0	0	0	0.46	0	0	0	0	0	0	0
MEFP	ME	0	0	0	3.36	0	0	0	0	0	0	0
PBSI	ME	0	0	0	16.3	0	0	0	0	0	0	0
MEBB	ME	0	0	0	2.31	0	0	0	0	0	0	0
MEKN	ME	0	0	0	9.60	0	1.74	0	0	0	0	0
MSPP	ME	0	0	0	0.98	0	0	0	0	0	19.0	0
MERY	ME	0	0	0	0.30	0	0	0	0	0	0	46.7
MEPR	ME	0	0	0	17.5	0	0.35	0	0	0	0	0
MEPH	ME	0	0	0	17.4	0	0	0	0	0	0	0
MESA	ME	0	0	0	12.3	0	0	0	0	0	0	0
CAKP	ME	0	0	0	13.4	0	0	0	0	0	0	0
MECC	ME	0	0	0	2.29	0	1.87	0	0	0	0	0
NHDP	NH	0	0	0	0.99	0	1.89	0	0	0	0	0
NHNM	NH	0	0	0	3.47	0	1.89	0	0	0	0	0
NHLH	NH	0	0	0	1.94	0	0	0	0	0	0	0
NHSM	NH	0	0	0	0.28	0	1.98	0	0	0	0	0
NHNC	NH	0	0	0	2.82	0	0	4.91	0	0	0	0
NHPI	NH	0	0	0	2.27	0	0	0	378	0	0	0
NHHS	NH	0	1.27	0	1.67	0	1.79	0	0	0	0	0
MAME	MA	3.75	0	0	3.22	0	0.97	0	0	0	0	0
SHFP	MA	0	0	0	3.06	0	0.42	0	0	0	0	0
MAPR	MA	0	0	15.10	0	4.17	0.58	0	0	0	0	0
MADI	MA	0	0	0	1.02	0	0	0	0	0	0	0
BHDI	MA	0	0	0	1.46	0	0.23	0	0	0	0	0
MABI	MA	0	0	0	0.84	0	0	0	0	59.4	0	0
MANR	MA	0	0	0	1.54	0	0.83	0	27.7	0	0	0
BHDB	MA	0	0	0	2.38	0	0	0	63.0	0	0	0
BHHB	MA	0	0	0	1.16	0	0	0	0	0	0	0
MAWR	MA	0	0	0	1.30	0	0	0	0	0	0	0
MACO	MA	0	0	0	2.62	0	0	0	0	0	0	0
MBNR	MA	0	0	0	31.0	0	0	0	0	0	0	0
MAMF	MA	0	0	0	3.19	0	0	0	0	0	0	0
DBCI	MA	0	0	0	2.78	0	0	0	0	0	0	0
BBCC	MA	0	0	0	6.98	0	0	0	0	0	0	0
MASN	MA	0	0	0	0	0	0	0	0	0	0	0
CCNH	MA	0	0	0	0.62	0	0	0	0	0	0	0

Appendix 4 (cont.). PPCP compound concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	Miconazole	Norgestrel	Propoxyphene	Propranolol	Ranitidine	Sertraline	Testosterone	Triamterene	Triclocarban
Minimum MDL		0.12	0.49	0.05	0.37	0.07	0.07	0.49	0.90	0.38
NSDI	NS	0	0.67	0	0	0	0	0	0	0
NSAR	NS	0	0	0	0	0	0	0	0	0
MEUR	ME	0	0	0	0	1.63	0	0	0	0
PBPI	ME	4.22	0	0	0	16.8	0	0	0	0
MEPI	ME	0	0	0	0	0	0	0	0	0
MEFP	ME	0	0	0	0	0	0	0	0	0
PBSI	ME	0	0	0	0	4.12	0	0	0	0
MEBB	ME	0	0	6.80	0	0	6.54	0	0	0
MEKN	ME	0	0	0	0	2.08	2.73	0	0	0
MSPP	ME	0	0	0	0	0	0	7.11	0	0
MERY	ME	0	0	0	0	0	2.05	0	0	0
MEPR	ME	0	0	0	0	1.64	3.67	0	0	0
MEPH	ME	0	0	0	0	0	14.2	0	0	0
MESA	ME	0	0	0	0	1.78	0	0	0	0
CAKP	ME	0	0	0	0	0	0	0	0	0
MECC	ME	0	0	0	0	0	2.29	0	1.25	0
NHDP	NH	0	0	0	42.6	0	3.41	0	2.19	0
NHNM	NH	2.92	0	0	0	0	3.75	0	0	0
NHLH	NH	0	0	0	0	0	0	0	0	0
NHSM	NH	0	0	0	0	0	3.48	0	1.71	0
NHNC	NH	0	0	0	0	0	1.74	0	0	0
NHPI	NH	0	0	0	27.7	0	0	0	2.45	0
NHHS	NH	0	0	0	0	0	1.69	0	0	0
MAME	MA	0	0	0	0	0	11.6	0	0	0
SHFP	MA	0	0	0	0	0	4.86	0	0	0
MAPR	MA	0	0	0	0	0	3.26	0	0	0
MADI	MA	0	0	0	0	0	4.20	12.4	0	0
BHDI	MA	0	0	0	0	0	3.03	10.9	0	1.09
MABI	MA	0	0	0	0	0	0	0	0	0
MANR	MA	0	0	0	0	0	0	0	1.82	5.38
BHDB	MA	0	0	0	0	0	2.40	0	0	4.37
BHHB	MA	0	0	0	0	2.49	0	0	0	0
MAWR	MA	0	0	0	0	0	0	0	0	0
MACO	MA	0	0	0	0	0	0	0	0	0
MBNR	MA	0	0	0	0	1.58	0	0	0	0
MAMF	MA	0	0	0	0	0	0	0	0	0
DBCI	MA	0	0	0	0	0	0	0	0	0
BBCC	MA	0	0	0	0	1.69	0	0	0	0
MASN	MA	0	0	0	0	0	0	0	0	0
CCNH	MA	0	0	0	0	0	0	0	0	0

Appendices

Appendix 5. PBDE compound concentrations (ng/wet g) above MDL measured in mussel tissue. Sites are listed geographically from north to south, following the coastline.

Site	Jurisdiction	PBDE-1	PBDE-47	PBDE-66	PBDE-71/49	PBDE-77	PBDE-99	PBDE-100	PBDE-118	PBDE-119	PBDE-126	PBDE-197	PBDE-209
Minimum MDL		0.04	0.05	0.03	0.04	0.05	0.04	0.09	0.04	0.04	0.04	0.02	0.34
NSDI	NS	0	0	0	0	0	0	0.22	0	0.07	0	0	0
NSFI	NS	0	0	0	0	0.51	0	0.29	0	0.09	0.07	0	0
NSAR	NS	0	0	0	0	0.13	0	0.26	0	0.07	0.07	0	0
MEUR	ME	0	0	0	0	0	0	0	0	0	0	0	0
PBPI	ME	0	0	0	0	0	0	0	0	0	0	0	0
MEPI	ME	0	0.08	0	0	0	0.09	0	0	0.11	0.27	0	0
MEFP	ME	0	0.43	0	0.27	0	0.20	0.19	0	0.08	0	0	0
PBSI	ME	0	0.24	0	0.10	0	0.12	0	0	0	0	0	0
MEBB	ME	0	0.16	0	0.21	0.09	0.11	0	0	0.12	0	0	0
MEKN	ME	0	0.09	0	0	0.09	0	0	0	0	0	0	0
MSPP	ME	0	0	0	0	0.13	0	0	0	0.12	0	0	0
MERY	ME	0	0.11	0	0	0.18	0.08	0	0	0.10	0	0	0
MEPR	ME	0	0.41	0	0.12	0	0.26	0	0	0	0	0	0
MEPH	ME	0	0.39	0	0.76	0	0.22	0	0	0	0	0	0
MESA	ME	0	0.18	0	0.06	0	0.11	0	0	0	0	0	0
CAKP	ME	0	0.08	0	0	0	0	0	0	0	0	0	0
MECC	ME	0	0.09	0	0.14	0.09	0	0	0	0.12	0	0	0
NHDP	NH	0	0.16	0	0.22	0.15	0.10	0	0	0.15	0	0	0
NHNM	NH	0	0.16	0	0.54	0.13	0.14	0	0	0.12	0	0	0
NHLH	NH	0	0.09	0	0.06	0.09	0	0	0	0.13	0	0	0
NHSM	NH	0	0.15	0	0.41	0	0.11	0	0	0	0	0	0
NHNC	NH	0	0.13	0	0.07	0.11	0.09	0	0	0.14	0	0	0
NHPI	NH	0	0.11	0	0.12	0	0.08	0	0	0.13	0	0	0
NHHS	NH	0	0.13	0	0	0.67	0.10	0	0.11	0.18	0.08	0	0
MAME	MA	0	0.37	0	0.08	0.12	0.19	0.16	0.08	0.08	0.07	0	1.04
SHFP	MA	0	0.31	0	0.08	0	0.13	0	0	0	0	0	0
MAPR	MA	0	0.49	0.06	0.29	0	0.26	0.20	0	0	0	0	0
MADI	MA	0	0.27	0	0.15	0.11	0.12	0	0	0	0	0	0
BHDI	MA	0	0.28	0	0.20	0	0.11	0	0	0	0	0	0
MABI	MA	0.46	0.20	0	0.08	0	0.09	0	0	0.13	0	0	0
MANR	MA	0	0.40	0.08	0.44	0.11	0.21	0.25	0	0	0.09	0	0
BHDB	MA	0	0.29	0.08	0.34	0	0.13	0.18	0	0	0	0	0
BHHB	MA	0	0.20	0.06	0.25	0	0.08	0	0	0	0	0	0
MAWR	MA	0	0.15	0.05	0.21	0.20	0.10	0	0	0.15	0	0	0
MACO	MA	0	0.12	0	0	0.18	0	0	0	0.14	0	0	0.96
MBNR	MA	0	0.11	0	0	0	0	0	0	0	0	0	0
MAMF	MA	0	0.09	0	0	0.38	0.09	0	0	0.16	0	0	0
DBCI	MA	0	0.11	0	0.15	0.07	0.07	0	0	0.13	0	0.05	0
BBCC	MA	0	0	0	0	0	0	0	0	0	0	0	0
MASN	MA	0	0.20	0	0	0.32	0	0	0	0.08	0	0	0
CCNH	MA	0	0	0	0	0	0	0	0	0	0	0	0

Appendix 6. Percent impervious surface and land-use category statistical results for site-based compound group detection frequencies and contaminant concentrations. Statistics were only tested when at least 12 (~30%) of the sites had detects. D, Developed; U, Undeveloped; O, Open-water.

Compound	Percent Impervious Surface					Land-use Classification				
	1 Km	2 Km	3 Km	4 Km	5 Km	1 Km	2 Km	3 Km	4 Km	5 Km
Total detection frequencies	p=.012 R^2=0.14	p=.005 R^2 =0.18	p=.002 R^2=0.21	p<.001 R^2=0.27	p<.001 R^2=0.30	p=.067	p=.045 D>O	p=.160	p=.064	p=.025 D>O
AP detection frequencies	p=.017 rho=0.39	p=.022 rho=0.37	p=.031 rho=0.35	p=.028 rho=0.36	p=.015 rho=0.39	p=.042 D>O D>U	p=.191	p=.279	p=.107	p=.108
PFAS detection frequencies	p=.015 rho=0.39	p=.088	p=.165	p=.190	p=.240	p=.127	p=.054	p=.088	p=.160	p=.301
PPCP detection frequencies	p<.001 rho=0.53	p=.001 rho=0.51	p=.003 rho=0.46	p=.001 rho=0.50	p<.001 rho=0.55	p=.015 D>O D>U	p=.185	p=.438	p=.047 D>U	p=.066
PBDE detection frequencies	p=.523	p=.369	p=.249	p=.121	p=.087	p=.522	p=.077	p=.088	p=.731	p=.336
NP1EO concentration	p=.014 rho=0.39	p=.018 rho=0.38	p=.021 rho=0.37	p=.019 rho=0.38	p=.010 rho=0.41	p=.022 D>O D>U	p=.105	p=.149	p=.058	p=.054
PFOSA concentration	p=.022 rho=0.37	p=.073	p=.128	p=.157	p=.201	p=.142	p=.032 D>O	p=.049 D>O	p=.081	p=.134
DEET concentration	p=.765	p=.746	p=.777	p=.793	p=.745	p=.252	p=.656	p=.553	p=.553	p=.513
Diphenhydramine concentration	p=.006 rho=0.44	p=.010 rho=0.41	p=.015 rho=0.39	p=.005 rho=0.44	p=.003 rho=0.47	p=.032 D>O D>U	p=.368	p=.652	p=.120	p=.183
Sertraline concentration	p=.002 rho=0.49	p=.011 rho=0.41	p=.034 rho=0.34	p=.015 rho=0.39	p=.005 rho=0.44	p=.152	p=.666	p=.721	p=.307	p=.215
PBDE-47 concentration	p=.100	p=.017 rho=0.38	p=.029 rho=0.36	p=.016 rho=0.39	p=.006 rho=0.43	p=.278	p=.163	p=.112	p=.300	p=.040 D>U
PBDE-71/49 concentration	p=.001 rho=0.51	p<.001 rho=0.60	p<.001 rho=0.57	p<.001 rho=0.57	p<.001 rho=0.59	p=.064	p=.034 D>O D>U	p=.035 D>O D>U	p=.002 D>O D>U	p<.001 D>O D>U
PBDE-77 concentration	p=.766	p=.564	p=.752	p=.916	p=.793	p=.476	p=.683	p=.910	p=.172	p=.233
PBDE-99 concentration	p=.011 rho=0.41	p=.006 rho=0.44	p=.010 rho=0.41	p=.007 rho=0.43	p=.002 rho=0.48	p=.094	p=.137	p=.046 D>U	p=.056	p=.002 D>O D>U
PBDE-119 concentration	p=.418	p=.199	p=.408	p=.377	p=.294	p=.498	p=.791	p=.563	p=.484	p=.557

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